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(54) Title: NOVEL HERBICIDALLY, ACARICIDALLY AND INSECTICIDALLY ACTIVE COMPOUNDS

$$\begin{array}{c} R_2 \\ N \\ R_3 \end{array} \stackrel{O}{\underset{O}{\longrightarrow}} R_1 \quad (1)$$

$$(R_4)_n \times (R_4)_n$$

$$(R_5)_m \times (R_5)_m$$

$$(a) (b) (c)$$

$$-x - (R_6)_0$$
 $-x - (R_6)_0$ (e)

(57) Abstract

The present invention relates to herbicidally, acaricidally and insecticidally active pyrazolidine-3,5-diones of formula (I), in which R₁ is (a), (b) or (c); R₂ and R₃ independently of one another are C₁-C₆alkyl; C₃-C₆alkenyl; or C₃-C₆alkynyl; or R₂ and R_3 together are a -(CH₂)₃-, -(CH₂)₄-, -CH₂-CH = CH-CH₂-, -CH₂-CH = CH- or -(CH₂)₂-CH = CH- bridge which is unsubstituted or up to trisubstituted by C₁-C₄alkyl; n is 0, 1, 2, 3 or 4; m is 0 or 1; the total of m and n being less than, or equal to, 4; the R₄ radicals independently of one another are halogen, nitro, cyano, C₁-C₄alkyl, C₁-C₄haloalkyl, C₁-C₁₀alkoxy, C_1 - C_4 alkoxy, C_3 - C_6 alkenyloxy, C_1 - C_4 alkoxy- C_2 - C_4 alkoxy, C_3 - C_6 alkynyloxy, C_1 - C_4 alkylcarbonyl, C_1 - C_4 alkylsulfinyl, C_1 - C_4 - C_4 - C_4 - C_4 - C_4 - C_4 - $C_$ (e); X is oxygen, sulfur, CH2 or NR7; o is 0, 1, 2 or 3; R6 radicals independently of one another are C1-C4alkyl, halogen, C₁-C₄haloalkyl, C₁-C₄haloalkoxy, C₁-C₄alkoxy, nitro, cyano, C₁-C₄alkoxycarbonyl, amino, mono-C₁-C₄alkylamino, or di-C₁-C₄alkylamino, and R₇ is hydrogen, C₁-C₄alkyl, formyl, or C₁-C₄alkylcarbonyl; the acid addition salts thereof, as well as processes for their preparation, and novel intermediates for these processes. The invention furthermore relates to herbicidally, acaricidally or insecticidally active compositions as well as to methods for controlling weeds, Acarina or insects,

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Novel herbicidally, acaricidally and insecticidally active compounds

The present invention relates to herbicidally, acaricidally and insecticidally active pyrazolidine-3,5-diones of the formula I, to processes for their preparation, and to novel intermediates for these processes. The invention furthermore relates to herbicidally, acaricidally or insecticidally active compositions as well as methods for controlling weeds. Acarina or insects.

The compounds according to the invention are those of the formula I

$$R_2$$
 R_3
 R_1
 R_3
 R_1
 R_1
 R_1
 R_1
 R_1
 R_2
 R_1
 R_1
 R_2
 R_1
 R_2
 R_1
 R_2
 R_1
 R_2
 R_1
 R_2
 R_3

in which

$$R_1$$
 is $(R_4)_n$; $(R_5)_m$; or $(R_5)_m$

 R_2 and R_3 independently of one another are C_1 - C_6 alkyl; C_3 - C_6 alkenyl; or C_3 - C_6 alkynyl; or R_2 and R_3 together are a -(CH₂)₃-, -(CH₂)₄-, -CH₂-CH=CH-CH₂-, -CH₂-CH=CH- or -(CH₂)₂-CH=CH- bridge which is unsubstituted or up to trisubstituted by C_1 - C_4 alkyl;

n is 0; 1; 2; 3; or 4;

m is 0; or 1; the total of m and n being less than, or equal to, 4; the

radicals independently of one another are halogen; nitro; cyano, C₁-C₄alkyl; C₁-C₄haloalkyl; C₁-C₁₀alkoxy; C₁-C₄haloalkoxy; C₃-C₆alkenyloxy; C₁-C₄alkoxy-C₂-C₄alkoxy; C₃-C₆alkynyloxy; C₁-C₄alkylcarbonyl;

 C_1 - C_4 alkoxycarbonyl; C_1 - C_4 alkylthio; C_1 - C_4 alkylsulfinyl; C_1 - C_4 alkylsulfonyl; amino; mono- C_1 - C_4 alkylamino; di- C_1 - C_4 alkylamino;

$$R_5$$
 is $-x$ $(R_6)_0$; $-x$ $(R_6)_0$

X is oxygen; sulfur; CH₂; or NR₇;

o is 0; 1; 2; or 3;

R₆ radicals independently of one another are C₁-C₄alkyl; halogen; C₁-C₄haloalkyl; C₁-C₄haloalkoxy; C₁-C₄alkoxy; nitro; cyano; C₁-C₄alkoxycarbonyl; amino; mono-C₁-C₄alkylamino; or di-C₁-C₄alkylamino; and

 R_7 is hydrogen; C_1 - C_4 alkyl; formyl; or C_1 - C_4 alkylcarbonyl; or the acid addition salts thereof.

Suitable acids for forming such acid addition salts are both organic and inorganic acids. Examples of such acids are, inter alia, hydrochloric acid, hydrobromic acid, nitric acid, various phosphoric acids, sulfuric acid, acetic acid, propionic acid, butyric acid, valeric acid, oxalic acid, malonic acid, maleic acid, fumaric acid, lactic acid, tartaric acid or salicylic acid.

Because of their chemical constitution, the compounds of the formula I can exist in the tautomeric equilibrium forms I = I' = I'':

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_2 \\ R_3 \\ \end{array}$$

$$\begin{array}{c} R_1 \\ R_3 \\ \end{array}$$

Moreover, certain substituents R_1 to R_7 , on their own or in combination with each other or in combination with the skeleton to which they are bonded, can have centres of chirality.

The invention extends to the racemate as well as to the enriched and optically pure forms of the stereoisomers in question.

In the processes described in the present application, the asymmetrically substituted compounds of the formula I are generally obtained in the form of racemates, unless chiral educts are used. The stereoisomers can then be isolated by methods known per se, such as fractional crystallisation following salt formation with optically pure bases, acids or metal complexes, or, alternatively, by chromatographic methods on the basis of their physicochemical properties.

The compounds of the formula I in which the radicals R_2 and R_3 are alkyl, alkenyl or alkynyl radicals are derivatives of the pyrazolidine-3,5-dione system. In those cases in which R_2 and R_3 are a saturated or partially unsaturated C_4 -carbon bridge, formula I is based on the ring system of the 1H-pyrazolo[1,2-a]pyridazine, and in those cases in which R_2 and R_3 are a saturated or partially unsaturated C_3 -carbon bridge, it is based on the ring system of 1H,5H-pyrazolo[1,2-a]pyrazole. The individual ring positions are numbered analogously to Chemical Abstracts:

1H-pyrazolo[1,2-a]pyridazine

1H,5H-pyrazolo[1,2-a]pyrazole

Halogen in the above definitions is to be understood as meaning fluorine, chlorine, bromine and iodine, preferably fluorine and chlorine.

Monoalkylamino is, in particular, methylamino, ethylamino, n-propylamino, i-propylamino and the isomeric butylamino radicals.

Dialkylamino within the given limits of the definition is the radical which is substituted by identical as well as different alkyl radicals; in particular dimethylamino, methylamino, diethylamino, dibutylamino and diisopropylamino.

Alkyl is methyl, ethyl, isopropyl, n-propyl, n-butyl, iso-butyl, sec-butyl, tert-butyl, as well

as the isomeric pentyl and hexyl radicals.

Examples of haloalkyl are fluoromethyl, difluoromethyl, trifluoromethyl, chloromethyl, dichloromethyl, trichloromethyl, 2,2,2-trifluoroethyl, 2-fluoroethyl, 2-chloroethyl and 2,2,2-trichloroethyl; preferably the halogen-substituted methyl radicals such as difluorochloromethyl, trifluoromethyl, difluoromethyl and dichlorofluoromethyl.

Alkoxy is methoxy, ethoxy, propyloxy, i-propyloxy, n-butyloxy, i-butyloxy, s-butyloxy and t-butyloxy; preferably methoxy and ethoxy.

Examples of alkoxyalkoxy are methoxyethoxy, ethoxyethoxy, propoxyethoxy, isopropoxypropoxy or tert-butoxybutoxy.

Examples of haloalkoxy are fluoromethoxy, difluoromethoxy, trifluoromethoxy, 2,2,2-trifluoroethoxy, 1,1,2,2-tetrafluoroethoxy, 2-fluoroethoxy, 2-chloroethoxy and 2,2,2-trichloroethoxy; preferably the halogen-substituted methoxy radicals such as difluoromethoxy and trifluoromethoxy.

Alkylthio is methylthio, ethylthio, propylthio, isopropylthio, n-butylthio, i-butylthio, s-butylthio, t-butylthio; preferably methylthio and ethylthio.

Alkylsulfinyl is methylsulfinyl, ethylsulfinyl, propylsulfinyl, isopropylsulfinyl, n-butylsulfinyl, i-butylsulfinyl, s-butylsulfinyl, t-butylsulfinyl; preferably methylsulfinyl and ethylsulfinyl.

Alkylsulfonyl is methylsulfonyl, ethylsulfonyl, propylsulfonyl, isopropylsulfonyl, n-butylsulfonyl, i-butylsulfonyl, s-butylsulfonyl, t-butylsulfonyl; preferably methylsulfonyl and ethylsulfonyl.

Alkenyl is to be understood as meaning straight-chain or branched alkenyl, such as allyl, methallyl, but-2-en-1-yl, pentenyl or 2-hexenyl. The alkenyl radicals are preferably bonded to the nitrogen hetero atom via a saturated carbon atom.

Alkynyl is to be understood as meaning straight-chain or branched alkynyl, such as propargyl, 1-methylprop-2-ynyl, but-2-yn-1-yl, or the isomeric pentynyl and 2-hexynyl radicals. The alkynyl radicals are preferably bonded to the nitrogen hetero atom via a

saturated carbon atom.

Alkylcarbonyl is, in particular, acetyl and propionyl.

Alkoxycarbonyl is methoxycarbonyl, ethoxycarbonyl, propyloxycarbonyl, i-propyloxycarbonyl, n-butyloxycarbonyl, i-butyloxycarbonyl, s-butyloxycarbonyl and t-butyloxycarbonyl; preferably methoxycarbonyl and ethoxycarbonyl.

In those substituents which are composed of a plurality of basic elements the individual elements may be selected freely within the limits of the definition.

Preferred compounds of the formula I

$$R_2$$
 N R_1 R_1 R_3 N Q R_1 Q Q

are those in which

$$R_1$$
 is $(R_4)_n$; $(R_4)_n$; or $(R_4)_n$; or

n is 0; 1; 2; 3; or 4;

m is 0; or 1; and the total of m and n is less than, or equal to, 4:

n' is 0; 1; 2; or 3;

n" is 0; 1; or 2;

m' is 0; or 1; and the total of m' and n' is less than, or equal to, 3; and the radicals R_1 to R_5 are as defined above.

Other preferred compounds are pyrazolidine-3,5-diones of the formula I

$$R_2$$
 R_3
 R_1
 R_3
 R_1
 R_1
 R_1
 R_1

in which

$$R_1$$
 is $(R_4)_n$ or $(R_5)_m$

 R_2 and R_3 independently of one another are C_1 - C_6 alkyl; C_3 - C_6 alkenyl; or C_3 - C_6 alkynyl; or R_2 and R_3 together are a -(CH₂)₃-, -(CH₂)₄-, -CH₂-CH=CH-CH₂-, -CH₂-CH=CH- or -(CH₂)₂-CH=CH- bridge which is unsubstituted or up to trisubstituted by C_1 - C_4 alkyl;

n is 0; 1; 2; 3; or 4;

m is 0; or 1; the total of m and n being less than, or equal to, 4; the

R₄ radicals independently of one another are halogen; nitro; cyano; C_1 - C_4 alkyl; C_1 - C_4 haloalkyl; C_1 - C_1 0alkoxy; C_1 - C_4 haloalkoxy; C_3 - C_6 alkenyloxy; C_3 - C_6 alkynyloxy; C_1 - C_4 alkylcarbonyl; C_1 - C_4 alkoxycarbonyl; C_1 - C_4 alkylsulfinyl; C_1 - C_4 alkylsulfonyl; amino; mono- C_1 - C_4 alkylamino; di- C_1 - C_4 alkylamino;

$$R_5$$
 is $-x$

X is oxygen; sulfur; CH₂; or NR₇;

o is 0; 1; 2; or 3;

 $R_6 \qquad \text{radicals independently of one another are C_1-C_4alkyl; halogen;} \\ C_1$-$C_4$haloalkyl;$C_1$-$C_4$haloalkoxy;$C_1$-$C_4$alkoxy; nitro; cyano;} \\ C_1$-$C_4$alkoxycarbonyl; amino; mono-C_1-C_4alkylamino; or di-C_1-C_4alkylamino; and is hydrogen; C_1-C_4alkyl; formyl; or C_1-C_4alkylcarbonyl,} \\$

where a preferred meaning in this group is that of the compounds of the formula I

$$R_2$$
 R_3
 R_1
 R_3
 R_1
 R_1
 R_1
 R_1
 R_1
 R_2
 R_3

- 7 -

in which

$$R_1$$
 is $(R_4)_m$ or $(R_5)_m$

n is 0; 1; 2; or 3;

m is 0; or 1; and the total of m and n is less than, or equal to, 4;

n' is 0; 1; 2; or 3;

m' is 0; or 1; and the total of m' and n' is less than, or equal to, 3; and the radicals R_2 to R_5 are as defined above.

In particular, the present invention relates to:

5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-diones of the formula Ia

$$(R_8)_p$$
 R_1
 R_1
 R_1
 R_1
 R_1
 R_1
 R_1
 R_1
 R_2
 R_1
 R_2
 R_3

in which

R₁ is as defined above;

 R_8 is C_1 - C_4 alkyl; and

p is 0, 1, 2 or 3, preferably 0;

5.8-dihydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-diones of the formula lb

$$(R_8)_p$$
 R_1 (Ib)

in which

R₁ is as defined above;

 R_8 is C_1 - C_4 alkyl; and

p is 0, 1, or 2, preferably 0;

7,8-dihydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-diones of the formula Ic

$$(R_8)$$
 $\stackrel{N}{=}$ $\stackrel{N}{=$

in which

R₁ is as defined above;

R₈ is C₁-C₄alkyl; and

p. is 0, 1, 2 or 3, preferably 0;

6.7-dihydro-1H.5H-pyrazolo[1,2-a]pyrazole-1,3(2H)-diones of the formula Id

$$(R_8)_p$$
. (Id)

in which

R₁ is as defined above;

 R_x is C_1 - C_4 alkyl; and

p is 0, 1, 2 or 3, preferably 0;

111.5H-pyrazolo[1,2-a]pyrazole-1,3(2H)-diones of the formula Ie

$$(R_g)_p$$
 N
 R_1 (Ie)

in which

R₁ is as defined above;

 R_s is C_1 - C_4 alkyl; and

p is 0, 1, 2 or 3, preferably 0;

pyrazolidine-1,3-diones of the formula If

in-which

 R_1 is as defined above; and R_2 and R_3 independently of one another are C_1 - C_6 alkyl; C_3 - C_6 alkenyl; or C_3 - C_6 alkynyl;

pyrazolidine-1,3-diones of the formula Ig

$$R_2$$
 R_1 R_3 R_1 R_1 (Ig),

in which

 R_1 is as defined above; and R_2 and R_3 independently of one another are C_1 - C_6 alkyl; or C_3 - C_6 alkenyl; and

pyrazolidine-1,3-diones of the formula Ih

$$R_2$$
 N R_1 (Ih),

in which

 R_1 is as defined above; and R_2 and R_3 are C_1 - C_6 alkyl.

Particularly preferred compounds are those of the formula I or of the formulae Ia to Ih in which

$$R_1$$
 is $N = (R_5)_m$ and R_5 is 0; 1; 2; or 3;

m' is 0; or 1; and the total of m' and n' is less than, or equal to, 3;

R₄ is not more than three times halogen; or C₁-C₄alkyl; not more than twice C₁-C₄alkoxy; C₁-C₄haloalkoxy; C₁-C₄alkylthio; C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino; mono-C₁-C₄alkylamino; di-C₁-C₄alkylamino; or C₁-C₄haloalkyl; and not more than once nitro; cyano; C₁-C₄alkylcarbonyl; C₁-C₄alkoxycarbonyl;

OL

$$R_1$$
 is $(R_4)_n$;

n is 0; 1; 2; 3; or 4;

m is 0; or 1; and the total of m and n is less than, or equal to, 4;

is not more than four times halogen; or C₁-C₄alkyl;
not more than three times C₁-C₁₀alkoxy; C₁-C₄haloalkoxy; or C₁-C₄alkylthio; and
not more than twice nitro; C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino;
mono-C₁-C₄alkylamino; di-C₁-C₄alkylamino; C₁-C₄haloalkyl; or cyano;
not more than once C₁-C₄alkylcarbonyl; C₃-C₆alkenyloxy; C₃-C₆alkynyloxy;
C₁-C₄alkoxycarbonyl; C₁-C₄alkoxy-C₂-C₄alkoxy; and

R₂, R₃ and R₅ are as defined above, and the meaning of the substituent R₄ can in each case be identical or different.

Compounds of the formula I or of the formulae Ia to Ih which must be emphasised are those in which

$$R_1$$
 is $N = \begin{pmatrix} R_4 \end{pmatrix}_m$ and $R_5 \end{pmatrix}_m$

n' is 0; 1; 2; or 3;

m' is 0;

OF

R₄ is not more than three times halogen; or C₁-C₄alkyl; not more than twice C₁-C₄alkoxy; C₁-C₄haloalkyl; C₁-C₄haloalkoxy; C₁-C₄alkylthio; C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino; mono-C₁-C₄alkylamino; or di-C₁-C₄alkylamino; and not more than once nitro; cyano; C₁-C₄alkylcarbonyl; C₁-C₄alkoxycarbonyl;

$$R_1$$
 is $(R_4)_n$

n is 0; 1; 2; or 3;

m is 0; or 1; and the total of m and n is less than, or equal to, 3;

is not more than three times fluorine; chlorine; or C₁-C₄alkyl; not more than twice C₁-C₄alkoxy; C₁-C₄haloalkyl; C₁-C₄haloalkoxy; or C₁-C₄alkylthio; and not more than once nitro; C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino; mono-C₁-C₄alkylamino; di-C₁-C₄alkylamino; cyano; C₁-C₄alkylcarbonyl; C₃-C₆alkenyloxy; C₃-C₆alkynyloxy; or C₁-C₄alkoxycarbonyl; and

 R_2 , R_3 and R_5 are as defined above, and the meaning of the substituent R_4 can in each case be identical or different.

Other compounds which must be emphasised are those of the formulae Ia to Ih in which

$$R_1$$
 is $N = \begin{pmatrix} (R_4)_n \\ (R_5)_m \end{pmatrix}$ and

n' is 0; 1; 2; or 3;

m' is 0;

R₄ is not more than three times fluorine; chlorine; or C₁-C₂alkyl; not more than twice C₁-C₂alkoxy; C₁-C₂haloalkyl; C₁-C₂haloalkoxy; C₁-C₂alkylthio; C₁-C₂alkylsulfinyl; C₁-C₂alkylsulfonyl; amino; mono-C₁-C₂alkylamino; or di-C₁-C₂alkylamino; and not more than once nitro; cyano; C₁-C₂alkylcarbonyl; C₁-C₂alkoxycarbonyl;

or

$$R_1$$
 is $(R_4)_m$

n is 0; 1; 2; or 3;

m is 0; or 1; and the total of m and n is less than, or equal to, 3;

R₄ is not more than three times fluorine; chlorine; or C_1 - C_4 alkyl; not more than twice C_1 - C_2 alkoxy; C_1 - C_2 haloalkyl; C_1 - C_2 haloalkoxy; or C_1 - C_2 alkylthio; and not more than once nitro; C_1 - C_2 alkylsulfinyl; C_1 - C_2 alkylsulfonyl; amino;

 $\label{eq:continuous} \mbox{mono-} C_1\text{-}C_2\mbox{alkylamino; di-}C_1\text{-}C_2\mbox{alkylamino; cyano; } C_1\text{-}C_2\mbox{alkylamino; cyano; } C_1\text{-}C_2\mbox{alkylamino; and}$

 R_2 , R_3 and R_5 are as defined above, and the meaning of the substituent R_4 can in each case be identical or different.

Further preferred compounds with regard to the insecticidal and acaricidal action are the following pyrazolidine-3,5-diones of the formula I

in which the following groups can be R₁

in which R_2 , R_3 , R_4 , R_5 , m and n have the abovementioned meaning and R_9 is halogen, C_1 - C_4 alkyi or C_1 - C_4 haloalkyl, the total m + n being less than, or equal to, 3.

In this context, pyrazolidine-3,5-diones of the formula I which must be particularly emphasised are those in which R_2 is methyl and R_3 is methyl or ethyl, or R_2 and R_3 together are -(CH₂)₃-, -(CH₂)₄- or -CH₂-CH-CH-CH₂.

CH₃CH₃

Pyrazolidine-3,5-diones of the formula I which must be particularly emphasised are furthermore those in which R_1 can be the following group:

$$R_1$$
 is R_{10} or 2-naphthyl

in which

R₉ is halogen; C₁-C₄alkyl; C₁-C₄haloalkyl;

R₁₀ is hydrogen; halogen; C₁-C₄alkyl; C₁-C₄haloalkyl

 R_{11} is hydrogen; halogen or C_1 - C_4 alkyl.

Particularly important in this sub-group are the pyrazolidine-3,5-diones, in which R_2 is methyl, R_3 is methyl or ethyl, or R_2 and R_3 together are -(CH₂)₃-, -(CH₂)₄- or -CH₂-CH-CH-CH₂ and R_1 is 2-naphthyl or CH₃CH₃

in which,

R₉ is chlorine; C₁-C₂alkyl; C₁-C₂haloalkyl;

R₁₀ is hydrogen; chlorine; fluorine; C₁-C₂alkyl or C₁-C₂haloalkyl; and

R₁₁ is hydrogen; fluorine; chlorine or methyl.

Individual compounds which may be mentioned are:

2-(phenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione (Comp. No. 1.001),

2 (2-methylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione (Comp. No. 1.002),

2-(4-methylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione (Comp. No. 1.003),

2-(2,4,6-trimethylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione (Comp. No. 1.010),

2-(4-chlorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione

(Comp. No. 1.013),

2-(2,4-dichlorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione (Comp. No. 1.014) and

2-(2,6-dichlorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione (Comp. No. 1.015),

2-(2,4-dimethylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione, (Comp. No. 1.004),

2-(2-chlorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione, (Comp. No. 1.012),

2-(2-chloro-6-fluorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione, (Comp. No. 1.016),

1,2-dimethyl-4-(2,4,6-trimethylphenyl)-3,5-pyrazolidinedione, (Comp. No. 10.010) in particular 2-(2,4,6-Trimethylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]-pyridazine-1,3(2H)-dione (Comp. No. 1.010).

The compounds of the formula I are novel. They can be prepared by

a) cyclisation of a hydrazinecarboxylate of the formula II,

in which R₁, R₂ and R₃ are as defined above and Y is C₁-C₆alkyl, phenyl or benzyl;

b) condensation of a malonic acid derivative of the formula III, in which R_1 is as defined above, with a hydrazine derivative of the formula IV, in which R_2 and R_3 are as defined above.

and in which Y is OH, halogen or C₁-C₄alkoxy; or

c) the reaction of a pyrazolidine-3,5-dione of the formula XXXIV

in which the radicals R_2 and R_3 are as defined above, with a compound of the formula XXXV

$$X-R_1$$
 (XXXV),

in which X and R_1 are as defined above, in the presence of a base or, if desired, in the presence of Cu(I) or of a Pd catalyst.

Reactions a), b), and c) are carried out analogously to processes known from the literature (N.R. El-Rayyes in Synthesis, 1985, 1028 et seq.), preferably in a solvent which is inert during the reaction. US 4 128 425 and J. Chem. Soc. Perkin Trans. I, 1987, 877 refer to processes for the preparation of the starting compounds (XXXIV).

Suitable bases for the cyclocondensation reaction a) are, in particular, sodium hydride, sodium amide, phenyllithium or potassium tert.-butylate.

The compounds of the formulae Ic and Ie,

$$(R_8)_{\overline{p}}$$
 $(R_8)_{\overline{p}}$
 $(R_8$

in which R_1 , R_8 and p are as defined above, can be prepared by reacting an alcohol of the formula XIII,

$$(R_8)_{p} \xrightarrow{OH} O \qquad R_1 \qquad H^+ /-H_2O \qquad x = 1$$

$$(XIII)$$
(Ie)

in which x is 0 or 1, in the presence of acid to give Ic or Ie.

A further process allows the pyrazolidine-1,3-diones of the formula If

$$R_2$$
 N
 R_3
 N
 R_1
 R_1 (If),

in which R_1 is as defined above and R_2 and R_3 independently of one another are C_1 - C_6 alkyl, C_3 - C_6 alkenyl or C_3 - C_6 alkynyl, to be obtained by acylating a hydrazone of the

formula XIV in which R₂ is as defined above and the radical $R_3 \subset C = is$

 C_1 - C_6 alkylidene, C_1 - C_6 alkenylidene or C_1 - C_6 alkynylidene radical, with a chloroformate IX in which Y is C_1 - C_4 alkyl, to give the N-acylhydrazone XV,

$$R_{2}$$
 NH
 R''_{3}
 N'
 R''_{3}
 R''_{3}

and subsequently acylating the N-acylhydrazone with arylacetyl halide of the formula X in which R_1 is as defined above and Z is chlorine or bromine, to give a hydrazine of the formula XVI,

and subsequently cyclising the hydrazine of the formula XVI in the presence of bases to give the pyrazolidine-1,3-dione of the formula If.

This process can be carried out analogously to the process known from EP-A-0 304 920 and Zh. Org. Khim. 4, (1968) p. 968.

The compounds of the formula II are valuable intermediates for the synthesis of the end products of the formula I according to the invention. The novel compounds of the formula II. processes for their preparation and novel starting compounds which are suitable for these processes are a further subject of the present invention.

The compounds of the formula II can be obtained by N-acylation of the N-acylhydrazines of the formula XVII in which R_2 and R_3 are as defined above and Y is C_1 - C_4 alkyl, with arylacetyl halides of the formula X in which R_1 is as defined above and Z is chlorine or bromine.

in analogy to processes known from the literature (Chem. Rev. 52 (1953), 237-416).

The compounds of the formula XVI

$$R_{2}$$
 CO-OY

 R_{3} $\stackrel{\dot{N}}{\sim}$ C CH_{2} R_{1}
 $\stackrel{\dot{C}}{\circ}$ (XVI)

in which R_1 is as defined above, Y is C_1 - C_4 alkyl and R_2 and R_3 independently of one another are C_1 - C_6 alkyl, C_3 - C_6 alkenyl or C_3 - C_6 alkynyl, can be prepared by acylating a hydrazone of the formula XIV in which R_2 is as defined above and the radical

R''₃

$$C = \text{is a } C_1\text{-}C_6\text{alkylidene, } C_1\text{-}C_6\text{alkenylidene or } C_1\text{-}C_6\text{alkynylidene radical,}$$

with a chloroformate IX in which Y is C₁-C₄alkyl, to give the N-acylhydrazone XV,

and subsequently acylating the N-acylhydrazone with an arylacetic halide of the formula X in which R_1 is as defined above and Z is chlorine or bromine, to give a hydrazine of the formula XVI

Alcohols of the formula XIII' can be prepared by hydrogenating a dihydropyridazin-(2H)3-one of the formula V, in which R_8 and p are as defined above, to give a tetrahydropyridazin-(2H)3-one VI and subsequently acylating the product with a chloroformate (IX), in which Y is C_1 - C_4 alkyl, to give a tetrahydropyridazin-(2H)3-one VII

$$(R_8)_{p} \xrightarrow{\text{NH}} (R_8)_{p} \xrightarrow{\text{NH}} (R_8)_{p} \xrightarrow{\text{CI-CO-OY}} (R_8)_{p} \xrightarrow{\text{NH}} (VII)$$

$$(VI) \qquad (VII)$$

and acylating the product obtained in this manner with an arylacetyl halide of the formula X in which R_1 is as defined above and Z is chlorine or bromine, to give a tetrahydropyridazin-(2H)3-one of the formula XI,

(VII) +
$$R_1 CH_2 CO-Z$$
 $(R_8)_p$ $(R_8)_p$ (XI)

then cyclising the tetrahydropyridazin-(2H)3-one XI in the presence of a base to give the pyrazolo[1,2-a]pyridazine XII and reducing this product with a hydrogenating agent, preferably sodium borohydride, to give the alcohol XIII'

(XII)
$$(R_8)_p \xrightarrow{O} (R_8)_p \xrightarrow{$$

The alcohols XIII'' can be obtained in an analogous manner by acylating a pyrazolidine-3-one (XVIII) in which R_8 and p are as defined above, with a chloroformate of the formula (IX) in which Y is C_1 - C_4 alkyl, to give the compound (XIX)

$$(R_8)_p \xrightarrow{\text{NH}} + CI \cdot CO \cdot OY \xrightarrow{(R_8)_p} \xrightarrow{\text{NH}} COOY$$

$$(XVIII) \qquad (XIX)$$

and acylating the product obtained in this manner with an arylacetyl halide of the formula X in which R_1 is as defined above and Z is chlorine or bromine, to give a pyrazolidin-3-one of the formula XX,

(XIX) +
$$R_1 CH_2 CO-Z$$

$$(R_8)_p \stackrel{O}{\underset{N-COOY}{|I|}} COOY$$
(XX)

then cyclising the diacylated pyrazolidin-3-one XX in the presence of a base to give the pyrazolo[1,2-a]pyrazole XXI and reducing this product with a hydrogenating agent, preferably sodium borohydride, to give the alcohol XIII",

(XXI) base
$$(R_8)_p$$
 (XXI) $(R_8)_p$ $(XIII''/x = 0)$

The N-acyl hydrazines of the formula XVII can be obtained by hydrolysis and decarboxylation of the hydrazinedicarboxylate XXII in which R_2 and R_3 are as defined above and Y is C_1 - C_4 alkyl,

The tetrahydropyridazinecarboxylates XXIII,

in which R_8 and p are as defined above and Y is C_1 - C_4 alkyl can furthermore be prepared by reacting a diene of the formula XXIV, in which R_8 and p are as defined above, with an azodicarboxylate of the formula XXV, in which Y is C_1 - C_4 alkyl

The compounds of the formula XXIII can subsequently be processed analogously to processes known from the literature (Coll. Czech. Chem. Commun. 33 (1968) 2087; Bull. Soc. Chim. France (1957) 704; EP-A-0 304 920 or Beilsteins Handbuch der Organischen Chemie [Manual of Organic Chemistry], Vol. 23^{III/IV}, 465) according to equation 1 below by hydrolysis and decarboxylation to give the tetrahydropyridazinecarboxylates XXVII, or by reduction, hydrolysis and decarboxylation via the diesters XXVII to give the hexahydropyridazines XXVIII.

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EQUATION 1

A further access route for the pyrazole- or pyridazinecarboxylates of the formula XXIX in which R_8 and p are as defined above and Y is C_1 - C_4 alkyl and n is 1 or 2, is the reaction of an α , ω -dihalo compound of the formula XXX in which R_8 and p are as defined above, Hal is halogen, preferably chlorine or bromine, and n is 1 or 2, with N,N'-hydrazinedicarboxylate XXXI in which Y is C_1 - C_4 alkyl, according to reaction equation 2:

EQUATION 2

$$(R_8)_p$$

$$(CH_2)_n + NH_{COOY}$$

$$(CH_2)_n N - COOY$$

$$(XXX) (XXXI) (XXXI)$$

The arylacetyl halides of the formula X are generally known or can be prepared from the corresponding arylacetic acids analogously to processes known from the literature. The following syntheses are particularly suitable for the preparation of the arylacetic acids:

- a) reaction of acetylaryl compounds in the sense of a Wilgerodt or Wilgerodt-Kindler reaction to give the corresponding arylacetamides, arylammonium salts or arylthioamides, followed by hydrolysis to give the arylacetic acids (E.V. Brown in Synthesis 1975, 358 et seq.).
- b) reaction in the sense of a Darzens reaction of suitably substituted arylaldehydes following Darzens method using using ethyl chloroacetate to give arylacetaldehydes, followed by oxidation to give the corresponding arylacetic acids (Ballester, Chem. Rev. <u>55</u> (1955) 283 et seq.).
- c) rearrangement of the α -haloalkyl aryl ketones XXXII or α -haloalkyl aryl ketals XXXIII; in which R_1 is as defined above and Hal is halogen, using processes known from the literature by zinc bromide catalysis (Synthesis, 1985, 436 or Angew. Chem. 1984, 413) to give the esters R_1CH_2COO-R ' which can subsequently be hydrolysed to the corresponding arylacetic acids.

$$R_{1} - CO - CH_{2} - Hal$$

$$(XXXII)$$

$$R_{1} - C (OR')_{2} - CH_{2} - Hal$$

$$(XXXIII)$$

$$R_{1} - C (XXXIII)$$

$$R_{1} - C (OR')_{2} - CH_{2} - Hal$$

$$(XXXIII)$$

$$R_{1} - CH_{2} - CO_{2} - R'$$

- d) hydrolysis of arylacetyl cyanides analogously to processes known from the literature (DE-A-3 416 772).
- e) reaction of aryl halides with malonic acid derivatives, phenylsulfonylacetonitriles or cyanoacetates analogously to processes known from the literature with Pd catalysis (Synthesis 1983, 67; Synthesis 1985, 506 or Chem. Lett. 1987, 887).

The compounds of the formula I are herbicidally active. As herbicides, the active ingredients of the formula I are generally used successfully at rates of application of 0.001 to 5 kg/ha, in particular 0.005 to 3 kg/ha. The dosage rate required for the desired action can be determined by experiments. It depends on the type of action, the development stage of the crop plant and of the weed, as well as on the application (location, time, method) and, due to these parameters, can vary within wide ranges.

When used at low rates of application, the compounds of the formula I are distinguished by growth-inhibiting and selectively herbicidal properties which make them outstandingly suitable for use in crops of useful plants, in particular in cereals, cotton, soybeans, rapeseed oil, maize and rice.

It has now been found that the compounds of the formula I according to the invention are valuable active ingredients in pest control while being well tolerated by warm-blooded species, fish and plants. The application of the active ingredients according to the invention particularly relates to insects and arachnids which can be found in useful plants and ornamentals in agriculture, in particular in cotton, vegetable and fruit crops, in the forest, in the protection of stored goods and materials as well as in the hygiene field, in particular on domestic animals and productive livestock. They are active against all or individual stages of development of normally sensitive, but also resistant, species. In this context, they may unfold their activity through immediate destruction of the pests or only after some time, for example during moulting, or through reduced oviposition and/or hatching rate. The abovementioned pests include:

from the order Lepidoptera, for example

Acleris spp., Adoxophyes spp., Aegeria spp., Agrotis spp., Alabama argillaceae, Amylois spp., Anticarsia gemmatalis, Archips spp., Argyrotaenia spp., Autographa spp., Busseola fusca, Cadra cautella, Carposina nipponensis, Chilo spp., Choristoneura spp., Clysia ambiguella, Cnaphalocrocis spp., Cnephasia spp., Cochylis spp., Coleophora spp., Crocidolomia binotalis, Cryptophlebia leucotreta, Cydia spp., Diatraea spp., Diparopsis castanea, Earias spp., Ephestia spp., Eucosma spp., Eupoecilia ambiguella, Euproctis spp., Euxoa spp., Grapholita spp., Hedya nubiferana, Heliothis spp., Hellula undalis, Hyphantria cunea, Keiferia lycopersicella, Leucoptera scitella, Lithocollethis spp., Lobesia botrana, Lymantria spp., Lyonetia spp., Malacosoma spp., Mamestra brassicae, Manduca sexta, Operophtera spp., Ostrinia nubilalis, Pammene spp., Pandemis spp., Panolis flammea, Pectinophora gossypiella, Phthorimaea operculella, Pieris rapae, Pieris spp., Plutella

xylostella, Prays spp., Scirpophaga spp., Sesamia spp., Sparganothis spp., Spodoptera spp., Synanthedon spp., Thaumetopoea spp., Tortrix spp., Trichoplusia ni and Yponomeuta spp.;

from the order of the Coleoptera, for example

Agriotes spp., Anthonomus spp., Atomaria linearis, Chaetocnema tibialis, Cosmopolites spp., Curculio spp., Dermestes spp., Diabrotica spp., Epilachna spp., Eremnus spp., Leptinotarsa decemlineata, Lissorhoptrus spp. Melolontha spp., Orycaephalus spp., Otiorhynchus spp., Phlyctinus spp., Popilia spp., Psylliodes spp., Rhizopertha spp., Scarabeidae, Sitophilus spp., Sitotroga spp., Tenebrio spp., Tribolium spp. and Trogoderma spp.;

from the order of the Orthoptera, for example

Blatta spp., Blattella spp., Gryllotalpa spp., Leucophaea maderae, Locusta spp.,

Periplaneta spp. and Schistocerca spp.;

from the order of the Isoptera, for example

Reticulitermes spp.;

from the order of the Psocoptera, for example

Liposcelis spp.;

from the order of the Anoplura, for example

Haematopinus spp., Linognathus spp. Pediculus spp., Pemphigus spp. and Phylloxera spp.;

from the order of the Mallophaga, for example

Damalinea spp. and Trichodectes spp.;

from the order of the Thysanoptera, for example

Frankliniella spp., Hercinothrips spp., Taeniothrips spp., Thrips palmi, Thrips tabaci and Scirtothrips aurantii;

from the order of the Heteroptera, for example

Cimex spp., Distantiella theobroma, Dysdercus spp., Euchistus spp. Eurygaster spp.

Leptocorisa spp., Nezara spp., Piesma spp., Rhodnius spp., Sahlbergella singularis,

Scotinophara spp. and Triatoma spp.;

from the order of the Homoptera, for example

Alcurothrixus floccosus. Aleyrodes brassicae, Aonidiella spp., Aphididae, Aphis spp.,

Aspidiotus spp., Bemisia tabaci, Ceroplaster spp., Chrysomphalus aonidium,

Chrysomphalus dictyospermi, Coccus hesperidum, Empoasca spp., Eriosoma larigerum,

Erythroneura spp., Gascardia spp., Laodelphax spp., Lecanium corni, Lepidosaphes spp.,

Macrosiphus spp., Myzus spp., Nephotettix spp., Nilaparvata spp., Paratoria spp.,

Pemphigus spp., Planococcus spp., Pseudaulacaspis spp., Pseudococcus spp., Psylla spp.,

Pulvinaria aethiopica, Quadraspidiotus spp., Rhopalosiphum spp., Saissetia spp.,

Scaphoideus spp., Schizaphis spp., Sitobion spp., Trialeurodes vaporariorum, Trioza erytreae and Unaspis citri;

from the order of the Hymenoptera, for example

Acromyrmex, Atta spp., Cephus spp., Diprion spp., Diprionidae, Gilpinia polytoma, Hoplocampa spp., Lasius spp., Monomorium pharaonis, Neodiprion spp., Solenopsis spp. and Vespa spp.;

from the order of the Diptera, for example

Aedes spp., Antherigona soccata, Bibio hortulanus, Calliphora erythrocephala, Ceratitis spp., Chrysomyia spp., Culex spp., Cuterebra spp., Dacus spp., Drosophila melanogaster, Fannia spp., Gastrophilus spp., Glossina spp., Hypoderma spp., Hyppobosca spp., Liriomyza spp., Lucilia spp., Melanagromyza spp., Musca spp., Oestrus spp., Orseolia spp. Oscinella frit, Pegomyia hyoscyami, Phorbia spp., Rhagoletis pomonella, Sciara spp., Stomoxys spp., Tabanus spp., Tannia spp. and Tipula spp.;

from the order of the Siphonaptera, for example

Ceratophyllus spp., Xenopsylla cheopis,

from the order of the Acarina, for example

Acarus siro, Aceria sheldoni, Aculus schlechtendali, Amblyomma spp., Argas spp., Boophilus spp., Brevipalpus spp., Bryobia praetiosa, Calipitrimerus spp., Chorioptes spp., Dermanyssus gallinae, Eotetranychus carpini, Eriophyes spp., Hyalomma spp., Ixodes spp., Olygonychus pratensis, Ornithodoros spp., Panonychus spp., Phyllocoptruta oleivora, Polyphagotarsonemus latus, Psoroptes spp., Rhipicephalus spp., Rhizoglyphus spp., Sarcoptes spp., Tarsonemus spp. and Tetranychus spp.; and from the order of the Thysanura, for example Lepisma saccharina.

The compounds are particularly suitable for controlling pests in cotton, fruit, rice and vegetable crops. Pests which are controlled are, in particular, those from the order Acarina, for example spider mites such as Tetranychus urticae and Panonychus ulmi, or ticks such as Boophilus spp..

As insecticides and acaricides, the active ingredients of the formula I are generally used at concentrations between 0.1 and 1000 ppm, preferably between 0.1 and 500 ppm. The rates of application per hectare are generally 1 to 2000 g of active ingredient per hectare, preferably 10 to 10000 g/ha, in particular 20 to 600 g/ha. The level of rates and concentrations of application for achieving an insecticidal and acaricidal pest control effect is much lower than is the case when these active ingredients are used as herbicides.

This means that damage of the treated useful plants is not possible when the active ingredients of the formula I are used as insecticides/acaricides within the teaching according to the invention.

The good pesticidal activity of the compounds of the formula I according to the invention corresponds to a mortality rate of at least 50-60 % of the abovementioned pests.

The effectiveness of the compounds according to the invention and of the compositions containing them can be substantially widened and adapted to the prevailing circumstances by adding other insecticides and/or acaricides. Examples of suitable additional substances are representatives from the following classes of active ingredients: organophosphorus compounds, nitrophenols and derivatives, formamidines, ureas, carbamates, pyrethroids, chlorinated hydrocarbons and Bacillus thuringiensis preparations.

The compounds of the formula I are used in unaltered form or, preferably, together with the auxiliaries conventionally used in the art of formulation, and they can therefore be processed in a known manner to give emulsifiable concentrates, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules and also encapsulations in polymeric substances. The methods of application such as spraying, atomising, dusting, scattering or pouring, as well as the compositions, are selected to suit the intended aims and the prevailing circumstances. The compounds of the formula I are futhermore also suitable for use in the treatment of seed. In this context, the seed can be treated or dressed with the active ingredient or with a formulation containing the active ingredient prior to sowing, and the active ingredient can be applied to the furrow during sowing.

i) Seed dressing

- a) The seeds are dressed with an active ingredient formulated as a wettable powder by shaking them in a container until the formulation is distributed evenly on the seed surface (dry dressing). Up to 4 g of active ingredient of the formula I are used (in the case of a 50 % formulation: up to 8.0 g of wettable powder) per kg of seed.
- b) The seeds are dressed with an emulsion concentrate of the active ingredient or with an aqueous solution of the active ingredient of the formula I which has been formulated as a wettable powder, following method a) (wet dressing).

c) The seed is dressed by being immersed for 1 to 72 hours in a liquor containing up to 1000 ppm of active ingredient of the formula I and, if appropriate, the seeds are then dried (seed soaking).

Naturally, seed dressing or treatment of the germinated seedling are preferred methods of application because the treatment with active ingredient is completely directed towards the target crop. As a rule, 4.0 g to 0.001 g of active substance are used per kg of seed, but it is possible to exceed, or remain under, the concentration limits given, depending on the method used, which also allows the addition of other active ingredients or micronutrients (repeated dressing).

Controlled release of active ingredients ii)

A solution of the active ingredient is applied to mineral granule carriers or polymerised granules (urea/formaldehyde) and allowed to dry. If appropriate, a coating can be applied (coated granules) which allows metered release of the active ingredient over a certain period.

The compounds of the formula I are employed in unaltered form or, preferably, as compositions together with the auxiliaries conventionally used in the art of formulation, and they are therefore processed in a known manner to give, for example, emulsion concentrates, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules, and also encapsulations, for example in polymeric substances. The application methods, such as spraying, atomising, dusting, scattering or pouring, as well as the type of composition are selected to suit the intended aims and the prevailing circumstances.

The formulations, i.e., the compositions, preparations or combinations containing the active ingredient of the formula I and, if desired, a solid or liquid additive, are prepared in a known manner, for example by intimately mixing and/or grinding the active ingredients with extenders, for example with solvents, solid carriers and, if desired, surface-active compounds (surfactants).

The following are possible as solvents: aromatic hydrocarbons, preferably the fractions C₈-C₁₂, for example xylene mixtures or substituted naphthalenes, phthalic esters, such as dibutyl phthalate or dioctyl phthalate, aliphatic hydrocarbons, such as cyclohexane or paraffins, alcohols and glycols as well as their ethers and esters, such as ethanol, ethylene glycol, ethylene glycol monomethyl ether or ethylene glycol monoethyl ether, ketones, such as cyclohexanone, strongly polar solvents, such as N-methyl-2-pyrrolidone, dimethyl sulfoxide or dimethylformamide, and also epoxidised or unepoxidised vegetable oils, such as epoxidised coconut oil or soya oil, or water.

Solid carriers which are generally used, for example for dusts and dispersible powders, are ground natural minerals, such as calcite, talc, kaolin, montmorillonite or attapulgite. To improve the physical properties, it is also possible to add highly-disperse silicas or highly-disperse absorptive polymers. Possible particulate, adsorptive carriers for granules are either porous types, for example pumice, brick grit, sepiolite or bentonite, or non-sorptive carrier materials, such as calcite or sand. Moreover, a large number of pregranulated materials of inorganic or organic nature can be used, such as, in particular, dolomite or comminuted plant residues.

Suitable surface-active compounds are non-ionic, cationic and/or anionic surfactants having good emulsifying, dispersing and wetting properties, depending on the nature of the active ingredient of the formula I to be formulated. Surfactants are also to be understood as meaning mixtures of surfactants.

Anionic surfactants which are suitable can be either so-called water-soluble soaps or water-soluble synthetic surface-active compounds.

Suitable soaps which may be mentioned are the alkali metal salts, alkaline earth metal salts or substituted or unsubstituted ammonium salts of higher fatty acids (C_{10} - C_{22}), such as the sodium salts or potassium salts of oleic or stearic acid, or of natural mixtures of fatty acids which can be obtained, for example, from coconut or tallow oil. Mention must also be made of the fatty acid methyltaurinates.

However, so-called synthetic surfactants are used more frequently, in particular fatty sulfonates, fatty sulfates, sulfonated benzimidazole derivatives or alkylarylsulfonates.

The fatty sulfates or fatty sulfonates are generally in the form of alkali metal salts, alkaline earth metal salts or substituted or unsubstituted ammonium salts, and generally have an alkyl radical having 8 to 22 C atoms, alkyl also including the alkyl moiety of acyl radicals, for example the sodium or calcium salt of ligninsulfonic acid, of the dodecylsulfuric ester or of a fatty alcohol sulfate mixture prepared from natural fatty acids. This group also

includes the salts of the sulfuric esters and sulfonic acids of fatty alcohol/ethylene oxide adducts. The sulfonated benzimidazole derivatives preferably contain 2 sulfonyl groups and one fatty acid radical having 8 to 22 C atoms. Examples of arylalkylsulfonates are the sodium, calcium or triethanolamine salts of dodecylbenzenesulfonic acid, of dibutylnaphthalenesulfonic acid or of a naphthalenesulfonic acid/formaldehyde condensation product.

Other suitable compounds are the corresponding phosphates, such as the salts of the phosphoric ester of a p-nonylphenol/(4-14)-ethylene oxide adduct or phospholipids.

Suitable non-ionic surfactants are mainly polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, saturated or unsaturated fatty acids and alkylphenols, which can contain 3 to 10 glycol ether groups and 8 to 20 carbon atoms in the (aliphatic) hydrocarbon radical and 6 to 18 carbon atoms in the alkyl radical of the alkylphenols.

Other non-ionic surfactants which are suitable are the water-soluble polyethylene oxide adducts with polypropylene glycol, ethylenediaminopolypropylene glycol and alkylpolypropylene glycol which have 1 to 10 carbon atoms in the alkyl chain and which comprise 20 to 250 ethylene glycol ether groups and 10 to 100 propylene glycol ether groups. The abovementioned compounds customarily comprise 1 to 5 ethylene glycol units per propylene glycol unit.

Examples of non-ionic surfactants which may be mentioned are nonylphenolpolyethoxyethanols, castor oil polyethylene glycol ethers, polypropylene/polyethylene oxide adducts, tributylphenoxypolyethoxyethanol, polyethylene glycol and octylphenoxypolyethoxyethanol.

Other suitable substances are fatty acid esters of polyoxyethylenesorbitan, such as polyoxyethylenesorbitan trioleate.

The cationic surfactants are mainly quaternary ammonium salts which contain at least one alkyl radical having 8 to 22 C atoms as N-substituent and which have lower halogenated or free alkyl, benzyl or lower hydroxyalkyl radicals as further substituents. The salts are preferably in the form of halides, methylsulfates or ethylsulfates, for example stearyltrimethylammonium chloride or benzyldi(2-choroethyl)ethylammonium bromide.

The surfactants customary in the art of formulation are described, inter alia, in the following publications:

- -"McCutcheon's Detergents and Emulsifiers Annual", Mc Publishing Corp., Glen Rock, New Jersey, 1988.
- M. and J. Ash, "Encyclopedia of Surfactants", Vol. I-III, Chemical Publishing Co., New York, 1980-1981.
- Dr. Helmut Stache "Tensid-Taschenbuch" [Surfactants Guide]", Carl Hanser Verlag, Munich/Vienna 1981.

As a rule, the agrochemical preparations contain 0.1 to 95 %, in particular 0.1 to 80 %, of the active ingredient of the formula I, 1 to 99.9% of a solid or liquid additive and 0 to 25 %, in particular 0.1 to 25 %, of a surfactant.

In particular, preferred formulations have the following composition: (% = per cent by weight)

			**
Emulsifiable concentrates:			
Active ingredient:	1 to	20	%, 5 to 10 % being preferred
Surface-active agent:	5 to	30	
Liquid carrier:	50 to	94	%, preferably 70 to 85 %
Dusts			
Active ingredient:	0.1	to	10 %, preferably 0.1 to 1 %
Solid carrier:	99.9	to	90 %, preferably 99.9 to 99 %
Suspension concentrates:			
Active ingredient:	5	to	75 %, preferably 10 to 50 %
Water:	94	to	24 %, preferably 88 to 30 %
Surface-active agent:	. 1	to	40 %, preferably 2 to 30 %
Wettable powder:			
Active ingredient:	0.5	to	90 %, preferably 1 to 80 %
Surface-active agent:	0.5	to	20 %, preferably 1 to 15 %

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Solid carrier: 5 to 99 %, preferably 15 to 90 %

Granules:

Active ingredient: 0.5 to 30 %, preferably 3 to 15 % Solid carrier: 99.5 to 70 %, preferably 97 to 85 %

While concentrated compositions are often preferred as commercially available goods, the end user will normally use dilute compositions. The use forms can be diluted down to 0.001 % active ingredient. The rates of application are generally 0.001 to 5 kg of a.i./ha, preferably 0.005 to 3 kg of a.i./ha.

The compositions can also contain further additives, such as stabilisers, defoamers, viscosity regulators, binders, tackifiers and also fertilisers or other active ingredients for achieving specific effects.

The examples which follow illustrate the invention.

Preparation Examples

H.1.2-(2,4,6-trimethylphenylacetyl)-1-ethoxycarbonylhexahydropyridazine

To a solution of 11 g (70 mmol) of ethyl hexahydropyridazine-1-carboxylate and 10.8 ml (70 mmol) of triethylamine in 350 ml of diethyl ether there is added dropwise with stirring at 20-25°C a solution of 13.8 g (70 mmol) of mesityleneacetyl chloride in 100 ml of diethyl ether. The mixture is subsequently stirred for a further 3 hours at room temperature. Precipitated triethyleneamine hydrochloride is then filtered off with suction, and the filtrate is concentrated in vacuo and chromatographed with ethyl acetate/hexane (1:1) on silica gel.

20.1 g (90.5 %) of the title compound of the formula

are isolated.

H.2.2-(2,4,6-trimethylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione

3.69 g (88 mmol) of a 60 % suspension of sodium hydride in white oil are introduced into 75 ml of toluene. To this mixture there are added dropwise at room temperature 22.3 g (70 mmol) of a solution of

2-(2,4,6-trimethylphenylacetyl)-1-ethoxycarbonylhexahydropyridazine in 75 ml of toluene, and the mixture is heated at the boil for 6 hours. 10 ml of ethanol are then added dropwise with ice-cooling, the reaction mixture is evaporated to dryness in vacuo, and the residue is dissolved in 200 ml of 1N NaOH. The product is precipitated from the resulting solution by adding concentrated hydrochloric acid at 0°C. The crude product is purified by recrystallisation from chloroform/hexane.

8.9 g of the title compound (Compound No. 1.010) of the formula

are isolated in the form of crystals of m.p. 244 - 246°C.

The compounds of Tables 1 to 14 can be prepared analogously to the above examples and to the preparation processes described:

Table 1

Compounds of the formula

$$\bigcap_{N} \bigcap_{O} \bigcap_{R_{n}} R_{n}$$

Comp.		R_n		Phys. data (°C)
Nr.				
1.001		Н		m.p. 184-185
1.002		2-CH ₃		m.p. 172,5-173,5
1.003		4-CH ₃		m.p. 186-187
1.004		2-CH ₃	4-CH ₃	m.p. 247-248
1.005		2-CH ₃	6-CH ₃	m.p. 209-210
1.006		2-CH ₃	5-CH ₃	m.p. 136-137
1.007		3-CH ₃	5-CH ₃	
1.008		2-CH ₃	3-CH ₃	
1.009		3-CH ₃	4-CH ₃	
1.010	2-CH ₃	4-CH ₃	6-CH ₃	m.p. 244-246
1.011	2-CH ₃	4-CH ₃	5-CH ₃	
1.012		2-Cl		m.p. 178-178,5
1.013		4-Cl		m.p. 206-207
1.014		2-Cl	4-Cl	m.p. 203-204
1.015		2-Cl	6-Cl	m.p. >250
1.016		2-CI	6-F	m.p. 218-219
1.017		2-CH ₃	4-Cl	
1.018		2-CH ₃	4-F	
1.019		2-Cl	4-CH ₃	
1.020		2-Cl	6-CH ₃	
1.021		2-F	4-F	,
1.022		2-F	6-F	m.p. 228-229
1.023		2-CH ₃	4-O-CH ₃	
1.024		2-CH ₃	6-O-CH ₃	
1.025		2-Cl	4-O-CH ₃	
1.026		2-Cl	6-O-CH ₃	

Comp. No.	· :	R _n	Phys. data (°C)
1.027	3-OCH ₃	4-OCH₃	
1.028	2-OCH ₃	5-OCH ₃	
1.029	2-OCH ₃	4-OCH ₃	
1.030	2-OCH ₃	6-OCH ₃	
1.031	2-CF ₃	6-CF ₃	
1.032	2-CF ₃	4-CF ₃	
1.033	3-CF ₃	5-CF ₃	
1.034	2-C1	4-CF ₃	m.p. 195-197
1.035	2-Cl	6-CF ₃	·
1.036	2-NO ₂	4-NO ₂	
1.037	2-Cl	4-NO ₂	
1.038	2-CH ₃	4-NO ₂	
1.039	2-O-CH ₃	4-NO ₂	
1.040	2-F	6-NO ₂	
1.041	2-Cl	6-NO ₂	
1.042	2-CH ₃	6-NO ₂	
1.043	2-O-CH ₃	6-NO ₂	•
1.044	2-F	4-NO ₂	
1.045	2-CH ₃	$4-N(C_2H_5)_2$	
1.046	2-C1	4-SO ₂ -CH ₃	
1.047	2-Cl	4-SO-CH ₃	
1.048	2-Cl	4-S-CH ₃	
1.049	2-Cl	6-SO ₂ -CH ₃	
1.050	2-Cl	6-SO-CH ₃	
1.051	2-Cl	6-S-CH ₃	
1.052	2-CH ₃	4-SO ₂ -CH ₃	
1.053	2-CH ₃	4-SO-CH ₃	
1.054	2-CH ₃	4-S-CH ₃	•
1.055	2-CH ₃	6-SO ₂ -CH ₃	
1.056	2-CH _{3.}	6-SO-CH ₃	
1.057	2-CH ₃	6-S-CH ₃	•
1.058	2-O-CH ₃	6-SO ₂ -CH ₃	•
1.059	2-O-CH ₃	6-SO-CH ₃	
1.060	2-O-CH ₃	6-S-CH ₃	•
1.061	2-O-CH ₃	4-SO ₂ -CH ₃	

1.062	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
1.065	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
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$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	·.
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
1.071	
1.072	
1.073	
1.074 2-Cl 4-CO-CH ₃ 1.075 2-O-CHF ₂ 4-O-CHF ₂ 1.076 2-Cl ₃ 4-O-CHF ₂ 1.077 2-Cl 4-O-CF ₃ 1.078 2-O-CF ₃ 4-O-CH ₃ 1.079 2-O-CHF ₂ 4-Cl 1.080 2-O-CHF ₂ 6-CH ₃ 1.081 2-O-CHF ₂ 6-Cl 1.082 2-O-CHF ₂ 4-CH ₃ 6-CH ₃ 1.083 2-CH ₃ 4-t-C ₄ H ₉ 6-CH ₃ 1.084 2-i-C ₃ H ₇ 4-i-C ₃ H ₇ 6-i-C ₃ H ₇ 1.085 2-CH ₃ 4-O-CH ₃ 6-CH ₃ 1.086 2-Cl 4-CF ₃ 6-Cl m.p. >260 1.087 2-Cl 4-CF ₃ 6-F	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
1.085 2-CH ₃ 4-O-CH ₃ 6-CH ₃ 1.086 2-Cl 4-CF ₃ 6-Cl m.p. >260 1.087 2-Cl 4-CF ₃ 6-F	
1.085 2-CH ₃ 4-O-CH ₃ 6-CH ₃ 1.086 2-Cl 4-CF ₃ 6-Cl m.p. >260 1.087 2-Cl 4-CF ₃ 6-F	
1.086 2-Cl 4-CF ₃ 6-Cl m.p. >260 1.087 2-Cl 4-CF ₃ 6-F	
1.087 2-Cl 4-CF ₃ 6-F	
1.088 2-Cl 4-NO ₂ 6-Cl	
1.089 2-Cl 4-Cl 6-Cl	
1.090 2-F 4-F 6-F	
1.()91 2-CH ₃ 4-NO ₂ 6-CH ₃	
1.092 2-Cl 4-Cl 6-CH ₃	
1.093 2-Cl 4-O-CH ₃ 6-Cl	
1.094 2-Cl 4-Cl 6-O-CH ₃	
1.095 2-F 4-O-CH ₃ 6-F	
1.096 2-O-CH ₃ 4-CH ₃ 6-O-CH ₃	

Comp. No.			R_n	Phys. data (°C)
1.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃	
1.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃	
1.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃	
1.100	2-CH ₃	4-Cl	6-CH ₃	
1.101	2-CH ₃	4-F	6-CH ₃	
1.102	2-CH ₃	4-CH ₃	6-O-CH ₃	•
1.103	2-F	4-Cl	5-O-i-C ₃ H ₇	m.p. 180-181
1.104	2-C1	4-Cl	5-O-CH ₃	
1.105		4-C1	5-O-CH ₃	
1.106	2-F	4-Cl	5-CO-O-CH ₃	
1.107	2-F	4-Cl	5-CO-O-C ₂ H ₅	
1.108		4-Cl	5-CO-O-CH ₃	
1.109	2-Cl	4-Cl	5-CO-O-i-C ₃ H ₇	
1.110		4 - 0		
1.111	••	4 - 0 - Cl		
1.112		4 - 0 — F	-	
1.113		4 - O - CF ₃		m.p. 178-179
1.114	2-CH ₃	4-0-		
1.115		4 - S —	·	
1.116		4 - S — CI		m.p. 242-243
1.117		4 - CH ₂		•
1.118		4 - CH ₂ — CI		

Comp.			R _n	Phys. data (°C)
1.119		4 - CH ₂ - F		
1.120		4 - CH ₂ - CF ₃		
1.121		4 - N-CHO		
1.122	2-F	4-Cl	5-O-CH ₂ -CH=CH ₂	
1.123	2-F	4-Cl	5-O-CH ₂ -C≡CH	
1.124	2-Br			m.p. 180-182
1.125	2-CF ₃			m.p. 185-186
1.126	2-OCH ₃	Ci		m.p. 191-194
1.127	2-CH ₃	4 .0 CI		
1.128	2-CH ₃	4-0-CF ₃		
1.129	2-CH ₃	4 -O -CI		
1.130	2-CH ₃	4-0-	6-CH ₃	
1.131	2-CH ₃	4-0-CF ₃	6-CH ₃	
1.132	2-CH ₃	4 -0 — CI	6-CH ₃	
1.133	2-CH ₃	4 -0 — C:	6-CH ₃	
1.134	2-CH ₃	Cl 4-Br	6-CH ₃	
1.135	2-CH ₃	6-C ₂ H ₅		
1.136	2-C ₂ H ₅	6-C ₂ H ₅		
1.137	2-CH ₃	4-OC ₂ H ₅	6-CH ₃	
1.138	2-CH ₃	4-O-i-C ₃ H ₇	6-CH ₃	
1.139	2-CH ₃	4-O-n-C ₃ H ₇	6-CH ₃	

Comp. No.			R _n	Phys. data (°C)
1.140	2-CH ₃	4-O-n-C ₁₀ H ₂₁	6-CH ₃	
1.141	2-CH ₃	4-O-n-C ₃ H ₇		·
1.142	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
1.143	2-CH ₃	4-O-(CH2)2OCH3	6-CH ₃	•
1.144	2-CH ₃	4-O-(CH2)2OCH3		
1.145	2-CH ₃	4-O-n-C ₆ H ₁₃	6-CH ₃	
1.146	2-CH ₃	4-0-		
1.147	2-CH ₃	4-0-N-	6-CH ₃	
1.148	2-CH ₃	4 -0 - CF ₃		
1.149	2-CH ₃	4 -O - CF ₃	6-CH ₃	
1.150	2-CH ₃	4 - O - CF ₃		
1.151	2-CH ₃	4 - O - CF ₃	6-CH ₃	
1.152	2-CH ₃	5 - 0 — CF ₃		
1.153	2-CH ₃	5 -O — CF ₃		
1.154	2-CH ₃	4 - 5 -	6-CH ₃	•
1.155	2-CII ₃	4 - s — CI	6-CH ₃	
1.156	2-C ₂ H ₅	4 - s —	6-CH ₃	

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Table 2

Compounds of the formula

$$\begin{array}{c|c}
 & \circ \\
 & \circ \\$$

Comp.	R_n		Phys. data (°C)
2.001	3-Cl		
2.002	3-F		
2.003	3-CH ₃		
2.004	5-Cl		
2.005	5-CF ₃		
2.006	3-Cl	5-Cl	
2.007	3-Cl	5-F	
2.008	3-Cl	5-CF ₃	m.p. 172-174
2.009	3-Cl	5-NO ₂	
2.010	3-Cl	5-SO ₂ -CH ₃	
2.011	3-F	5-F	
2.012	3-F	5-Cl	
2.013	3-F	5-CF ₃	
2.014	3-NO ₂	5-NO ₂	
2.015	3-NO ₂	5-Cl	
2.016	3-NO ₂	5-CF ₃	
2.017	3-CF ₃	5-C!	
2.018	3-CF ₃	5-CF ₃	
2.019	3-CH ₃	5-CH ₃	

Table 3

Compounds of the formula

$$\bigcap_{n=0}^{N} \bigcap_{n=0}^{R_n} R_n$$

Comp. No.	R _n		Phys. data (°C)
3.001	Н		
3.002	2-CH ₃		
3.003	4-CH ₃		
3.004	2-CH ₃	4-CH ₃	·
3.005	2-CH ₃	6-CH ₃	•
3.006	2-CH ₃	5-CH ₃	
3.007	3-CH ₃	5-CH ₃	
3.008	2-CH ₃	3-CH ₃	
3.009	3-CH ₃	4-CH ₃	
3.010 2-CH ₃	4-CH ₃	6-CH ₃	m.p. 191-193
3.011 2-CH ₃	4-CH ₃	5-CH ₃	••
3.012	2-Cl	·	
3.013	4-Cl		
3.014	2-Cl	4-Cl	
3.015	2-Cl	6-Cl	m.p. 213-215
3.016	2-CI	6-F	
3.017	2-CH ₃	4-C!	
3.018	2-CH ₃	4-F	
3.019	2-Cl	4-CH ₃	
3.020	2-Cl	6-CH ₃	
3.021	2-F	4-F	
3.022	2-F	6-F	
3.023	2-CH ₃	4-O-CH ₃	·.

Comp.	R_n	Phys. data (°C)	
3.024	2-CH ₃	6-O-CH ₃	
3.025	2-Cl	4-O-CH ₃	
3.026	2-Cl	6-O-CH ₃	
3.027	3-OCH ₃	4-OCH ₃	
3.028	2-OCH ₃	5-OCH ₃	
3.029	2-OCH ₃	4-OCH ₃	
3.030	2-OCH ₃	6-OCH ₃	
3.031	2-CF ₃	6-CF ₃	
3.032	2-CF ₃	4-CF ₃	•
3.033	3-CF ₃	5-CF ₃	
3.034	2-Cl	4-CF ₃	
3.035	2-Cl	6-CF ₃	
3.036	2-NO ₂	4-NO ₂	
3.037	2-Cl	4-NO ₂	
3.038	2-CH ₃	4-NO ₂	
3.039	2-O-CH ₃	4-NO ₂	
3.040	2-F	6-NO ₂	
3.041	2-Cl	6-NO ₂	
3.042	2-CH ₃	6-NO ₂	
3.043	2-O-CH ₃	6-NO ₂	
3.044	2-F	4-NO ₂	
3.045	2-CH ₃	$4-N(C_2H_5)_2$	
3.046	2-C1	4-SO ₂ -CH ₃	
3.047	2-Cl	4-SO-CH ₃	
3.048	2-Cl	4-S-CH ₃	
3.049	2-Cl	6-SO ₂ -CH ₃	
3.050	2-Cl	6-SO-CH ₃	
3.051	2-Cl	6-S-CH ₃	
3.052	2-CH ₃	4-SO ₂ -CH ₃	
3.053	2-CH ₃	4-SO-CH ₃	
3.054	2-CH ₃	4-S-CH ₃	
3.055	2-CH ₃	6-SO ₂ -CH ₃	
3.056	2-CH ₃	6-SO-CH ₃	
3.057	2-CH ₃	6-S-CH ₃	
3.058	2-O-CH ₃	6-SO ₂ -CH ₃	

Comp.		R_n		Phys. data (°C)
		•		
3.059		2-O-CH ₃	6-SO-CH ₃	
3.060		2-O-CH ₃	6-S-CH ₃	
3.061		2-O-CH ₃	4-SO ₂ -CH ₃	•
3.062		2-O-CH ₃	4-SO-CH ₃	
3.063		2-O-CH ₃	4-S-CH ₃	
3.064		2-CH ₃	$6-N(C_2H_5)_2$	
3.065		2-Cl	$6-N(CH_3)_2$	
3.066		2-Cl	$4-N(CH_3)_2$	
3.067		2-Cl	4-CO ₂ CH ₃	-
3.068		2-CH ₃	$6-CO_2C_2H_5$	
3.069		2-CH ₃	$4-CO_2C_2H_5$	
3.070		2-CH ₃	4-CN	
3.071	,	2-CH ₃	6-CN	
3.072		2-C1	4-CN	
3.073		2-Cl	6-CN	
3.074		2-Cl	4-CO-CH ₃	
3.075		2-O-CHF ₂	4-O-CHF ₂	
3.076		2-CH ₃	4-O-CHF ₂	
3.077		2-Cl	4-O-CF ₃	
3.078		2-O-CF ₃	4-O-CH ₃	
3.079		2-O-CHF ₂	4-Cl	
3.080		2-O-CHF ₂	6-CH ₃	
3.081		2-O-CHF ₂	6-Cl	
3.082	2-O-CHF ₂	4-CH ₃	6-CH ₃	
3.083	2-CH ₃	4-t-C ₄ H ₉	6-CH ₃	
3.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇	
3.085	2-CH ₃		6-CH ₃	· .
3.086	2-Cl	4-CF ₃	6-Cl	
3.087	2-C1	4-CF ₃	6-F	
3.088	2-Cl	4-NO ₂	6-Cl	
3.089	2-Cl	4-Cl	6-C1	
3.090	2-F	4-F	6-F	
3.091	2-CH ₃	4-NO ₂	6-CH ₃	
3.092	2-Cl	4-Cl	6-CH ₃	
3.093	2-Cl	4-O-CH ₃	6-Cl	

Comp.		R _n	Phys. data (°C)	
3.094	2-Cl	4-Cl	6-O-CH ₃	
3.095	2-F	4-O-CH ₃	6-F	
3.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃	
3.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃	
3.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃	
3.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃	
3.100	2-CH ₃	4-Cl	6-CH ₃	
3.101	2-CH ₃	4-F	6-CH ₃	
3.102	2-CH ₃	4-CH ₃	6-O-CH ₃	
3.103	2-F	4-Cl	5-O-i-C ₃ H ₇	
3.104	2-Cl	4-Cl	5-O-CH ₃	
3.105		4-Cl	5-O-CH ₃	
3.106	2-F	4-Cl	5-CO-O-CH ₃	
3.107	2-F	4-Cl	5-CO-O-C ₂ H ₅	
3.108		4-Cl	5-CO-O-CH ₃	
3.109	2-CI	4-Cl	5-CO-O-i-C ₃ H ₇	
3.110		4 - 0 -		
3.111		4 - O — CI		
3.112		4 - 0 — F		
3.113		4-0-CF ₃		
3.114	2-CH ₃	4-0-		
3.115		4 - S —		
3.116		4 - S — CI		
3.117		4 - CH ₂		

Comp.		R _n		Phys. data (°C)
3.118		4 - CH ₂ —CI		
3.119		4 - CH ₂ - F		
3.120		4 - CH ₂ - CF ₃		
3.121		4 - N-CHO		
3.122	2-F	4-Cl	5-O-CH ₂ -CH=CH ₂	
3.123	2-F	4-Cl	5-O-CH ₂ -C≡CH	
3.124	2-Br		2	
3.125	2-CF ₃			
3.126	2-OCH ₃			
3.127 3.128	2-CH ₃	4-0-Cl 4-0-CF ₃		
3.129	2-CH ₃	4 -0 -C1		
3.130	2-CH ₃	4-0-	6-CH ₃	
3.131	2-CH ₃	4 - O — CF ₃	6-CH ₃	
3.132	2-CH ₃	4 -0 -CI	6-CH ₃	
3.133	2-CH ₃	4 -0 -C1	6-CH ₃	
3.134 3.135	2-CH ₃ 2-CH ₃	6-C ₂ H ₅	6-CH ₃	

Comp.		R_n	Phys. data (°C)	
3.136	2-C ₂ H ₅	6-C ₂ H ₅		
3.137	2-CH ₃	4-OC ₂ H ₅	6-CH ₃	
3.138	2-CH ₃	4-O-i-C ₃ H ₇	6-CH ₃	
3.139	2-CH ₃	4-O-n-C ₃ H ₇	6-CH ₃	
3.140	2-CH ₃	4-O-n-C ₁₀ H ₂₁	6-CH ₃	
3.141	2-CH ₃	4-O-n-C ₃ H ₇		
3.142	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
3.143	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃	6-CH ₃	
3.144	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
3.145	2-CH ₃	4-O-n-C ₆ H ₁₃	6-CH ₃	
3.146	2-CH ₃	4-o-\(\bigve{N}\)	•	
3.147	2-CH ₃	4-0-	6-CH ₃	
3.148	2-CH ₃	4 -0 -CF ₃		
3.149	2-CH ₃	4 -0 - CF ₃	6-CH ₃	
3.150	2-CH ₃	4-0-KP3		
3.151	2-CH ₃	4 - O - CF ₃	6-CH ₃	
3.152	2-CH ₃	5-0-KP3		
3.153	2-CH ₃	5-0-CF ₃		
3.154	2-CH ₃	4 - S —	6-CH ₃	

Comp. No.	R _n .		Phys. data (°C)
3.155 2-CH ₃	4 - s — C1	6-CH ₃	
3.156 2-C ₂ H ₅	4 - S —	6-CH ₃	

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Table 4
Compounds of the formula

$$\bigcap_{CH_3}^{N} \bigcap_{O}^{R_n}$$

Comp. No.	R _n		Phys. data (°C)	_
4.001	Н			
4.002	2-CH ₃			
4.003	4-CH ₃			
4.004	2-CH ₃	4-CH ₃		
4.005	2-CH ₃	6-CH ₃		
4.006	2-CH ₃	5-CH ₃		
4.007	3-CH ₃	5-CH ₃		
4.008	2-CH ₃	3-CH ₃		
4.009	3-CH ₃	4-CH ₃		
4.010 2-CH ₃	4-CH ₃	6-CH ₃	m.p. 206-207	
4.011 2-CH ₃	4-CH ₃	5-CH ₃		
4.012	2-Cl			
4.013	4-Cl			
4.014	2-Cl	4-Cl		
4.015	2-Cl	6-Cl		
4.016	2-Cl	6-F		
4.017	2-CH ₃	4-Cl		
4.018	2-CH ₃	4-I-		
4.019	2-Cl	4-CH ₃		
4.020	2-Cl	6-CH ₃		
4.021	2-F	4-F		
4.022	2-F	6-F		
4.023	2-CH ₃	4-O-CH ₃		

Comp. No.		R_n	Phys. data (°C)
4.024	2-CH ₃	6-O-CH ₃	
4.025	2-C1	4-O-CH ₃	
4.026	2-C1	6-O-CH ₃	
4.027	3-OCH ₃	4-OCH ₃	
4.028	2-OCH ₃	5-OCH ₃	
4.029	2-OCH ₃	4-OCH ₃	
4.030	2-OCH ₃	6-OCH ₃	
4.031	2-CF ₃	6-CF ₃	
4.032	2-CF ₃	4-CF ₃	
4.033	3-CF ₃	5-CF ₃	
4.034	2-C1	4-CF ₃	
4.035	2-C1	6-CF ₃	
4.036	2-NO ₂	4-NO ₂	
4.037	2-Cl	4-NO ₂	
4.038	2-CH ₃	4-NO ₂	
4.039	2-O-CH ₃	4-NO ₂	•
4.040	2-F	6-NO ₂	• •
4.041	2-Cl	6-NO ₂	
4.042	2-CH ₃	6-NO ₂	
4.043	2-O-CH ₃	6-NO ₂	
4.044	2-F	4-NO ₂	
4.045	2-CH ₃	$4-N(C_2H_5)_2$	
4.046	2-Cl	4-SO ₂ -CH ₃	
4.047	2-Cl	4-SO-CH ₃	
1.048	2-Cl	4-S-CH ₃	
1.049	2-C1	6-SO ₂ -CH ₃	
1.050	2-Cl	6-SO-CH ₃	•
1.051	2-C1	6-S-CH ₃	
1.052	2-CH ₃	4-SO ₂ -CH ₃	
1.053	2-CH ₃	4-SO-CH ₃	
1.054	2-CH ₃	4-S-CH ₃	
.055	2-CH ₃	6-SO ₂ -CH ₃	
.056	2-CH ₃	6-SO-CH ₃	
.057	2-CH ₃	6-S-CH ₃	
.058	2-O-CH ₃	6-SO ₂ -CH ₃	· :

Comp No.		R_n	Phys. data (°C)
4.059		2-O-CH ₃	6-SO-CH ₃
4.060		2-O-CH ₃	6-S-CH ₃
4.061		2-O-CH ₃	4-SO ₂ -CH ₃
4.062		2-O-CH ₃	4-SO-CH ₃
4.063		2-O-CH ₃	4-S-CH ₃
4.064		2-CH ₃	$6-N(C_2H_5)_2$
4.065		2-Cl	6-N(CH ₃) ₂
4.066		2-Cl	4-N(CH ₃) ₂
4.067		2-Cl	4-CO ₂ CH ₃
4.068		2-CH ₃	$6-CO_2C_2H_5$
4.069		2-CH ₃	$4-CO_2C_2H_5$
4.070		2-CH ₃	4-CN
4.071		2-CH ₃	6-CN
4.072		2-Cl	4-CN
4.073		2-Cl	6-CN
4.074		2-CI	4-CO-CH ₃
4.075		2-O-CHF ₂	4-O-CHF ₂
4.076		2-CH ₃	4-O-CHF ₂
4.077		2-Cl	4-O-CF ₃
4.078		2-O-CF ₃	4-O-CH ₃
4.079		2-O-CHF ₂	4-Cl
4.080		2-O-CHF ₂	6-CH ₃
4.081		2-O-CHF ₂	6-Cl
4.082	2-O-CHF ₂	4-CH ₃	6-CH ₃
4.083	2-CH ₃	$4-t-C_4H_9$	6-CH ₃
4.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇
4.085	2-CH ₃	4-O-CH ₃	6-CH ₃
4.086	2-Cl	4-CF ₃	6-Cl
1.087	2-C1	4-CF ₃	6-F
4.088	2-Cl	4-NO ₂	6-Cl
1.089	2-Cl	4-Cl	6-Cl
1.090	2-F	4-F	6-F
1.091	2-CH ₃	4-NO ₂	6-CH ₃
1.092	2-Cl	4-Cl	6-CH ₃
1.093	2-Cl	4-O-CH ₃	6-Cl

Comp.			R_n	Phys. data (°C)
4.094	2-Cl	4-Cl	6-O-CH ₃	
4.095	2-F	4-O-CH ₃	6-F	
4.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃	
4.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃	
4.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃	
4.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃	-
4.100	2-CH ₃	4-Cl	6-CH ₃	
4.101	2-CH ₃	4-F	6-CH ₃	•
4.102	2-CH ₃	4-CH ₃	6-O-CH ₃	
4.103	2-F	4-Cl	5-O-i-C ₃ H ₇	
4.104	2-Cl	4-Cl	5-O-CH ₃	• •
4.105		4-Cl	5-O-CH ₃	
4.106	2-F	4-Cl	5-CO-O-CH ₃	
4.107	2-F	4-Cl	5-CO-O-C ₂ H ₅	
4.108		4-Cl	5-CO-O-CH ₃	
4.109	2-Cl	4-Cl	5-CO-O-i-C ₃ H ₇	
4.110		4-0-	·	
4.111		4 - 0 — CI		·
4.112		4 - 0 - F	-	
4.113		4-0-CF ₃		
4.114	2-CH ₃	4-0-		
4.115		4 - S		
4,116	.	4 - S — CI		
4.117	•	4 - CH ₂		

Comp.		R _n	Phys. data (°C)
4.118		4 - CH ₂ —CI	
4.119		4 - CH ₂ - F	
4.120		4 - CH ₂ - CF ₃	
4.121		4 - N-CHO	
4.122 4.123	2-F 2-F	4-Cl 4-Cl	5-O-CH ₂ -CH=CH ₂ 5-O-CH ₂ -C≡CH

Table 5

Compounds of the formula

Comp.		R _n	Phys. data (°C)
5.001		Н	
5.002		2-CH ₃	
5.003		4-CH ₃	
5.004		2-CH ₃	4-CH ₃
5.005		2-CH ₃	6-CH ₃
5.006		2-CH ₃	5-CH ₃
5.007		3-CH ₃	5-CH ₃
5.008		2-CH ₃	3-CH ₃
5.009		3-CH ₃	4-CH ₃
5.010	2-CH ₃	4-CH ₃	6-CH ₃
5.011	2-CH ₃	4-CH ₃	5-CH ₃
5.012		2-C1	
5.013		4-Cl	
5.014		2-Cl	4-Cl
5.015		2-Cl	6-Cl
5.016		2-Cl	6-F
5.017		2-CH ₃	4-Cl
5.018		2-CH ₃	4-F
5.019		2-Cl	4-CH ₃
5.020		2-Cl	6-CH ₃
5.021		2-F	4-F
5.022		2-F	6-F

Comp.	R_n	Phys. data (°C)	
5.023	2-CH ₃	4-O-CH ₃	
5.024	2-CH ₃	6-O-CH ₃	
5.025	2-Cl	4-O-CH ₃	
5.026	2-Cl	6-O-CH ₃	
5.027	3-OCH ₃	4-OCH ₃	
5.028	2-OCH ₃	5-OCH ₃	
5.029	2-OCH ₃	4-OCH ₃	
5.030	2-OCH ₃	6-OCH ₃	
5:031	2-CF ₃	6-CF ₃	
5.032	2-CF ₃	4-CF ₃	
5.033	3-CF ₃	5-CF ₃	
5.034	2-Cl	4-CF ₃	
5.035	2-Cl	6-CF ₃	
5.036	2-NO ₂	4-NO ₂	
5.037	2-Cl	4-NO ₂	
5.038	2-CH ₃	4-NO ₂	
5.039	2-O-CH ₃	4-NO ₂	
5.040	2-F	6-NO ₂	
5.041	2-Cl	6-NO ₂	
5.042	2-CH ₃	6-NO ₂	
5.043	2-O-CH ₃	6-NO ₂	
5.044	2-F	4-NO ₂	
5.045	2-CH ₃	$4-N(C_2H_5)_2$	
5.046	2-Cl	4-SO ₂ -CH ₃	
5.047	2-Cl	4-SO-CH ₃	
5.048	2-Cl	4-S-CH ₃	
5.049	2-Cl	6-SO ₂ -CH ₃	
5.050	2-Cl	6-SO-CH ₃	
5.051	2-Cl	6-S-CH ₃	
5.052	2-CH ₃	4-SO ₂ -CH ₃	
5.053	2-CH ₃	4-SO-CH ₃	
5.054	2-CH ₃	4-S-CH ₃	
5.055	2-CH ₃	6-SO ₂ -CH ₃	
5.056	2-CH ₃	6-SO-CH ₃	
5.057	2-CH ₃	6-S-CH ₃	

Comp.		R_n^{\cdot}	Phys. data (°C)	·
No.				
5.058		2-O-CH ₃	6-SO ₂ -CH ₃	
5.059		2-O-CH ₃	6-SO-CH ₃	
5.060		2-O-CH ₃	6-S-CH ₃	
5.061		2-O-CH ₃	4-SO ₂ -CH ₃	•
5.062		2-O-CH ₃	4-SO-CH ₃	
5.063		2-O-CH ₃	4-S-CH ₃	
5.064		2-CH ₃	$6-N(C_2H_5)_2$	
5.065		2-Cl	6-N(CH ₃) ₂	
5.066		2-Cl	4-N(CH ₃) ₂	
5.067		2-Cl	4-CO ₂ CH ₃	
5.068		2-CH ₃	$6-CO_2C_2H_5$	·
5.069		2-CH ₃	$4-CO_2C_2H_5$	
5.070		2-CH ₃	4-CN	
5.071		2-CH ₃	6-CN	
5.072		2-Cl	4-CN	
5.073		2-Cl	6-CN	
5.074		2-Cl	4-CO-CH ₃	
5.075		2-O-CHF ₂	4-O-CHF ₂	
5.076		2-CH ₃	4-O-CHF ₂	,
5.077		2-C1	4-O-CF ₃	
5.078		2-O-CF ₃	4-O-CH ₃	
5.079		2-O-CHF ₂	4-Cl	
5.080		2-O-CHF ₂	6-CH ₃	
5.081		2-O-CHF ₂	6-Cl	
5.082	2-O-CHF ₂	4-CH ₃	6-CH₃	
5.083	2-CH ₃	4-t-C₄H ₉	6-CH ₃	
5.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇	
5.085	2-CH ₃	4-O-CH ₃	6-CH ₃	•
5.086	2-Ci	4-CF ₃	6-Cl	
5.087	2-Cl	4-CF ₃	6-F-	
5.088	2-Cl	4-NO ₂	6-Cl	
5.089	2-C1	4-Cl	6-Cl	
5.090	2-F	4-F	6-F	_
5.091	2-CH ₃	4-NO ₂	6-CH ₃	
5.092	2-Cl	4-Cl	6-CH ₃	

Comp.		R _n	Phys. data (°C)
5.093	2-Cl	4-O-CH ₃	6-Cl
5.094	2-Cl	4-CI	6-O-CH ₃
5.095	2-F	4-O-CH ₃	6-F
5.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃
5.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃
5.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃
5.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃
5.100	2-CH ₃	4-Cl	6-CH ₃
5.101	2-CH ₃	4-F	6-CH ₃
5.102	2-CH ₃	4-CH ₃	6-O-CH ₃
5.103	2-F	4-Cl	5-O-i-C ₃ H ₇
5.104	2-Cl	4-Cl	5-O-CH ₃
5.105		4-Cl	5-O-CH ₃
5.106	2-F	4-Cl	5-CO-O-CH ₃
5.107	2-F	4-Cl	5-CO-O-C ₂ H ₅
5.108		4-Cl	5-CO-O-CH ₃
5.109	2-Cl	4-Cl	5-CO-O-i-C ₃ H ₇
5.110		4 - 0	
5.111		1 - 0 — CI	
5.112		4 - 0 — F	
5.113		4-0-CF3	-
5.114	2-CH ₃	4-0-	
5.115		4 - S —	
5.116		4 - S — CI	

Comp. No.		R _n	Phys. data (°C)
			
5.117		4 - CH ₂	
5.118		4 - CH ₂ —CI	
5.119		4 - CH ₂ F	
5.120		4 - CH ₂ - CF ₃	
5.121		4 - N CHO	
5.122	2-F	4-Cl	5-O-CH ₂ -CH=CH ₂
5 123	2-F	4-Cl	5-O-CH ₂ -C≡CH

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Table 6

Compounds of the formula

Comp.		R _n		Phys. data (°C)	
6.001		Н			
6.002		2-CH ₃			
6.003		4-CH ₃			
6.004		2-CH ₃	4-CH ₃		
6.005		2-CH ₃	6-CH ₃		• 9
6.006		2-CH ₃	5-CH ₃		
6.007		3-CH ₃	5-CH ₃		
6.008		2-CH ₃	3-CH ₃		·
6.009		3-CH ₃	4-CH ₃		
6.010	2-CH ₃	4-CH ₃	6-CH ₃	m.p. 224	
6.011	2-CH ₃	4-CH ₃	5-CH ₃		
6.012	•	2-Cl			
6.013		4-Cl			
6.014		2-Cl	4-Cl		
6.015		2-Cl	6-Cl		
6.016		2-Cl	6-F		
6.017		2-CH ₃	4-C1		
6.018		2-CH ₃	4-F		
6.019		2-Cl	4-CH ₃		
6.020		2-Cl	6-CH ₃		:
6.021		2-F	4-F		
6.022		2-F	6-F		

	•		•
Comp.	R_n		Phys. data (°C)
No.			· ·
6.023	2-CH ₃	4-O-CH ₃	
6.024	2-CH ₃	6-O-CH ₃	
6.025	2-C1	4-O-CH ₃	
6.026	2-Cl	6-O-CH ₃	
6.027	3-OCH ₃	4-OCH ₃	
6.028	2-OCH ₃	5-OCH ₃	
6.029	2-OCH ₃	4-OCH ₃	
6.030	2-OCH ₃	6-OCH ₃	
6.031	2-CF ₃	6-CF ₃	
6.032	2-CF ₃	4-CF ₃	
6.033	3-CF ₃	5-CF ₃	·
6.034	2-Cl	4-CF ₃	
6.035	2-Cl	6-CF ₃	
6.036	2-NO ₂	4-NO ₂	
6.037	2-Cl	4-NO ₂	
6.038	2-CH ₃	4-NO ₂	
6.039	2-O-CH ₃	4-NO ₂	
6.040	2-F	$6-NO_2$	
6.041	2-Cl	$6-NO_2$	
6.042	2-CH ₃	6-NO ₂	•
6.043	2-O-CH ₃	6-NO ₂	
6.044	- 2-F	4-NO ₂	
6.045	2-CH ₃	$4-N(C_2H_5)_2$	
6.046	2-Cl	4-SO ₂ -CH ₃	
6.047	·· 2-Cl	4-SO-CH ₃	·
6.048	2-Cl	4-S-CH ₃	
6.049	2-Cl	6-SO ₂ -CH ₃	
6.050	2-Cl	6-SO-CH ₃	
6.051	2-Cl	6-S-CH ₃	•
6.052	2-CH ₃	4-SO ₂ -CH ₃	·
6.053	2-CH ₃	4-SO-CH ₃	
6.054	2-CH ₃	4-S-CH ₃	
6.055	2-CH ₃	6-SO ₂ -CH ₃	
6.056	2-CH ₃	6-SO-CH ₃	
6.057	2-CH ₃	6-S-CH ₃	

Comp. No.	R _n	Phys. data (°C)
6.058	2-O-CH ₃	6-SO ₂ -CH ₃
6.059	2-O-CH ₃	6-SO-CH ₃
6.060	2-O-CH ₃	6-S-CH ₃
6.061	2-O-CH ₃	4-SO ₂ -CH ₃
6.062	2-O-CH ₃	4-SO-CH ₃
6.063	2-O-CH ₃	4-S-CH ₃
6.064	2-CH ₃	$6-N(C_2H_5)_2$
6.065	2-Cl	6-N(CH ₃) ₂
6.066	2-Cl	4-N(CH ₃) ₂
6.067	2-Cl	4-CO ₂ CH ₃
6.068	2-CH ₃	$6-CO_2C_2H_5$
6.069	2-CH ₃	$4-CO_2C_2H_5$
6.070	2-CH ₃	4-CN
6.071	2-CH ₃	6-CN
6.072	2-Cl	4-CN
6.073	2-Cl	6-CN
6.074	2-Cl	4-CO-CH ₃
6.075	2-O-CHF ₂	4-O-CHF ₂
6.076	2-CH ₃	4-O-CHF ₂
6.077	2-Cl	4-O-CF ₃
6.078	2-O-CF ₃	4-O-CH ₃
6.079	2-O-CHF ₂	4-Cl
6.080	2-O-CHF ₂	6-CH ₃
6.081	2-O-CHF ₂	6-Cl
6.082	2-O-CHF ₂ 4-CH ₃	6-CH ₃
6.083	2-CH ₃ 4-t-C ₄ H ₉	6-CH ₃
6.084	2-i-C ₃ H ₇ 4-i-C ₃ H ₇	6-i-C ₃ H ₇
6.085	2-CH ₃ 4-O-CH ₃	6-CH ₃
6.086	2-Cl 4-CF ₃	6-Cl
6.087	2-Cl 4-CF ₃	6-F
6.088	2-Cl 4-NO ₂	6-Cl
6.089	2-Cl 4-Cl	6-Cl
6.090	2-F 4-F	6-F
6.091	2-CH ₃ 4-NO ₂	6-CH ₃
6.092	2-Cl 4-Cl	6-CH ₃

Comp.		R_n	Phys. data (°C)		
No.					
6.093	2-Cl	4-O-CH ₃	6-Cl		
6.094	2-C1	4-Cl	6-O-CH ₃		
6.095	2-F	4-O-CH ₃	6-F		
6.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃		
6.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃		
6.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃		
6.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃		
6.100	2-CH ₃	4-C1	6-CH ₃		
6.101	2-CH ₃	4-F	6-CH ₃		
6.102	2-CH ₃	4-CH ₃	6-O-CH ₃		
6.103	2-F	4-Cl	5-O-i-C ₃ H ₇		
6.104	2-Cl	4-C1	5-O-CH ₃		
6.105		4-Cl	5-O-CH ₃		
6.106	2-F	4-Cl	5-CO-O-CH ₃		
6.107	2-F	4-Cl	5-CO-O-C ₂ H ₅		
6.108		4-C1	5-CO-O-CH ₃		
6.109	2-Cl	4-Cl	5-CO-O-i-C ₃ H ₇		
6.110		4 - 0			
6.111		4 - O — CI			
6.112		4 - 0 — F			
6.113		4 - O - CF ₃			
6.114	2-CH ₃	4 - 0 -			
6.115		4 - S -			
6.116		4 - S — CI			

Comp.		R _n .	Phys. data (°C)
6.117		4 - CH ₂	
6.118		4 - CH ₂ —C1	
6.119		4 - CH ₂ F	
6.120		4 - CH ₂ —CF ₃	
6.121		4 - N CHO	
6.122	2-F	4-Cl	5-O-CH ₂ -CH=CH ₂
6.123	2-F	4-Cl	5-O-CH ₂ -C≡CH
6.124	2-Br		
6.125	2-CF ₃		
6.126	2-OCH ₃	Cl	
6.127	2-CH ₃	4 -0 -CI	
6.128	2-CH ₃	4-0-CF ₃	
6.129	2-CH ₃	4 -0 — CI	
6.130	2-CH ₃	4-0-	6-CH ₃
6.131	2-CH ₃	4-0-CF ₃	6-CH ₃
6.132	2-CH ₃	4 -0 -CI	6-CH ₃
6.133	2-CH ₃	4 -0 — CI	6-CH ₃

Comp.		R_n	·	Phys. data (°C)
6.134	2-CH ₃	4-Br	6-CH ₃	
6.135	2-CH ₃	$6-C_2H_5$		
6.136	$2-C_2H_5$	$6-C_2H_5$		
6.137	2-CH ₃	4-OC ₂ H ₅	6-CH ₃	
6.138	2-CH ₃	4-O-i-C ₃ H ₇	6-CH ₃	
6.139	2-CH ₃	4-O-n-C ₃ H ₇	6-CH ₃	
6.140	2-CH ₃	4-O-n-C ₁₀ H ₂₁	6-CH ₃	
6.141	2-CH ₃	4-O-n-C ₃ H ₇		
6.142	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
6.143	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃	6-CH ₃	
6.144	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
6.145	2-CH ₃	4-O-n-C ₆ H ₁₃	6-CH ₃	· · · · · · · · · · · · · · · · · · ·
6.146	2-CH ₃	4-0-		
6.147	2-CH ₃	4 -o — N	6-CH ₃	
6.148	2-CH ₃	4 -O - CF ₃		
6.149	2-CH ₃	4 -O - CF ₃	6-CH ₃	
6.150	2-CH ₃	4-0-CF ₃		
6.151	2-CH ₃	CI N————————————————————————————————————	6-CH ₃	
6.152	2-CH ₃	5 - 0 — CF ₃		
6.153	2-CH ₃	5 - O - CF ₃		

Comp.		R _n		Phys. data (°C)
6.154	2-CH ₃	4 - S —	6-CH ₃	
6.155	2-CH ₃	4 - s — CI	6-CH ₃	
6.156	2-C ₂ H ₅	4 -s —	6-CH ₃	

Table 7

Compounds of the formula

Comp. No.	R _n		Phys. data (°C)
7.001	Н		
7.002	2-CH ₃		
7.003	4-CH ₃		
7.004	2-CH ₃	4-CH ₃	
7.005	2-CH ₃	6-CH ₃	
7.006	2-CH ₃	5-CH ₃	
7.007	3-CH ₃	5-CH ₃	
7.008	2-CH ₃	3-CH ₃	
7.009	3-CH ₃	4-CH ₃	
7.010 2-0	CH ₃ 4-CH ₃	6-CH ₃	
7.011 2-0	CH ₃ 4-CH ₃	5-CH ₃	
7.012	2-Cl		·*
7.013	4-Cl		
7.014	2-Cl	4-Cl	
7.015	2-Cl	6-Cl	
7.016	2-C1	6-F	
7.017	2-CH ₃	4-Cl	•
7.018	2-CH ₃	4-F	
7.019	2-Cl	4-CH ₃	
7.020	2-Cl	6-CH ₃	
7.021	2-F	4-F	
7.022	2-F	6-F	

Comp.	R_n	Phys. data (°C)
7.023	2-CH ₃	4-O-CH ₃
7.024	2-CH ₃	6-O-CH ₃
7.025	2-Cl	4-O-CH ₃
7.026	2-Cl	6-O-CH ₃
7.027	3-OCH ₃	4-OCH ₃
7.028	2-OCH ₃	5-OCH ₃
7.029	2-OCH ₃	4-OCH ₃
7.030	2-OCH ₃	6-OCH ₃
7.031	2-CF ₃	6-CF ₃
7.032	2-CF ₃	4-CF ₃
7.033	3-CF ₃	5-CF ₃
7.034	2-Cl	4-CF ₃
7.035	2-Cl	6-CF ₃
7.036	2-NO ₂	4-NO ₂
7.037	2-Cl	4-NO ₂
7.038	2-CH ₃	4-NO ₂
7.039	2-O-CH ₃	4-NO ₂
7.040	2-F	6-NO ₂
7.041	2-Cl	6-NO ₂
7.042	2-CH ₃	6-NO ₂
7.043	2-O-CH ₃	6-NO ₂
7.044	2-F	4-NO ₂
7.045	2-CH ₃	$4-N(C_2H_5)_2$
7.046	2-Cl	4-SO ₂ -CH ₃
7.047	2-Cl	4-SO-CH ₃
7.048	2-Cl	4-S-CH ₃
7.049	2-Cl	6-SO ₂ -CH ₃
7.050	2-C1	6-SO-CH₃
7.051	2-Cl	6-S-CH ₃
7.052	2-CH ₃	4-SO ₂ -CH ₃
7.053	2-CH ₃	4-SO-CH ₃
7.054	2-CH ₃	4-S-CH ₃
7.055	2-CH ₃	6-SO ₂ -CH ₃
7.056	2-CH ₃	6-SO-CH ₃
7.057	2-CH ₃	6-S-CH ₃
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			•	•
Comp.		R_n		Phys. data (°C)
No.		·		
7.058		2-O-CH ₃	6-SO ₂ -CH ₃	·
7.059		2-O-CH ₃	6-SO-CH ₃	
7.060		2-O-CH ₃	6-S-CH ₃	
7.061		2-O-CH ₃	4-SO ₂ -CH ₃	
7.062		2-O-CH ₃	4-SO-CH ₃	
7.063		2-O-CH ₃	4-S-CH ₃	
7.064		2-CH ₃	$6-N(C_2H_5)_2$	
7.065		2-Cl	6-N(CH ₃) ₂	
7.066		2-Cl	4-N(CH ₃) ₂	••
7.067		2-Cl	4-CO ₂ CH ₃	
7.068		2-CH ₃	$6-CO_2C_2H_5$	·
7.069		2-CH ₃	$4-CO_2C_2H_5$	
7.070		2-CH ₃	4-CN	
7.071		2-CH ₃	6-CN	
7.072		2-Cl	4-CN	
7.073		2-Cl	6-CN	
7.074		2-Cl	4-CO-CH ₃	
7.075		2-O-CHF ₂	4-O-CHF ₂	
7.076		2-CH ₃	4-O-CHF ₂	
7.077		2-C1	4-O-CF ₃	
7.078	•	2-O-CF ₃	4-O-CH ₃	
7.079		2-O-CHF ₂	4-Cl	
7.080		2-O-CHF ₂	6-CH ₃	
7.081		2-O-CHF ₂	6-Cl	
7.082	2-O-ÇHF ₂	4-CH ₃	6-CH ₃	
7.083	2-CH ₃	4-t-C ₄ H ₉	6-CH ₃	
7.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇	
7.085	2-CH ₃	4-O-CH ₃	6-CH ₃	
7.086	-	4-CF ₃	6-Cl	
7.087	2-Cl	4-CF ₃	6-F	
7.088	2-Cl	4-NO ₂	6-Cl	•
7.089	2-Cl	4-Cl	6-Cl	•
7.090	2-F	4-F	6-F	
7.091	2-CH ₃	4-NO ₂	6-CH ₃	
7.091	7-C113	7-1107	0-0117	

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Comp.		R_n	Phys. data (°C)
7.093	2-CI	4-O-CH ₃	6-Cl
7.094	2-Cl	4-Cl	6-O-CH ₃
7.095	2-F	4-O-CH ₃	6-F
7.096	2-O-CH ₃	4-CH ₃	6-O-CH₃
7.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃
7.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃
7.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃
7.100	2-CH ₃	4-Cl	6-CH ₃
7.101	2-CH ₃	4-F	6-CH ₃
7.102	2-CH ₃	4-CH ₃	6-O-CH ₃
7.103	2-F	4-Cl	5-O-i-C ₃ H ₇
7.104	2-Cl	4-Cl	5-O-CH ₃
7.105		4-Cl	5-O-CH ₃
7.106	2-F	4-Cl	5-CO-O-CH ₃
7.107	2-F	4-Cl	5-CO-O-C ₂ H ₅
7.108		4-Cl	5-CO-O-CH ₃
7.109	2-Cl	4-Cl	5-CO-O-i-C ₃ H ₇
7.110		4 - 0 -	
7.111		4 - 0 — CI	
7.112		4 - 0 — F	
7.113		4-0-CF3	
7.114	2-CH ₃	4-0-	
7.115		4 - S —	
7.116		4 - S — CI	

Comp. No.	R _n	Phys. data (°C)
7.117	4 - CH ₂	
7.118	4 - CH ₂ — CI	
7.119	4 - CH ₂ F	
7.120	4 - CH ₂ - CF ₃	• •
7.121	4 - N CHO	
7.122 2-F	4-Cl	5-O-CH ₂ -CH=CH ₂
7.123 2-F	4-Cl	5-O-CH ₂ -C≡CH

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Table 8

Compounds of the formula

$$\bigcap_{N} \bigcap_{O} \bigcap_{O} \bigcap_{R_{n}} \bigcap_{R_{n}} \bigcap_{R_{n}} \bigcap_{R_{n}} \bigcap_{C} \bigcap_{C}$$

Comp. No.	R_n	Phys. data (°C)	
8.001	Н		
8.002	2-CH ₃		
8.003	4-CH ₃		
8.004	2-CH ₃	4-CH ₃	
8.005	2-CH ₃	6-CH ₃	
8.006	2-CH ₃	5-CH ₃	
8.007	3-CH ₃	5-CH ₃	
8.008	2-CH ₃	3-CH ₃	
8.009	3-CH ₃	4-CH ₃	
8.010 2-	CH ₃ 4-CH ₃	6-CH ₃	
	CH ₃ 4-CH ₃	5-CH ₃	
8.012	2-Cl		
8.013	4-Cl		
8.014	2-Cl	4-Cl	
8.015	2-Cl	6-Cl	
8.016	2-Cl	6-F	
8.017	2-CH ₃	4-Cl	
8.018	2-CH ₃	4-F	
8.019	2-Cl	4-CH ₃	
8.020	2-Cl	6-CH ₃	
8.021	2-F	4-F	
8.022	2-F	6-F	

Comp. No.	R _n		Phys. data (°C)
8.023	2-CH ₃	4-O-CH ₃	
8.024	2-CH ₃	6-O-CH ₃	
8.025	2-Cl	4-O-CH ₃	
8.026	2-Cl	6-O-CH ₃	
8.027	3-OCH ₃	4-OCH ₃	
8.028	2-OCH ₃	5-OCH ₃	
8.029	2-OCH ₃	4-OCH ₃	
8.030	2-OCH ₃	6-OCH ₃	
8.031	2-CF ₃	6-CF ₃	
8.032	2-CF ₃	4-CF ₃	
8.033	3-CF ₃	5-CF ₃	
8.034	2-Cl	4-CF ₃	
8.035	2-C1	6-CF ₃	•
8.036	2-NO ₂	4-NO ₂	
8.037	2-Cl	4-NO ₂	
8.038	2-CH ₃	4-NO ₂	
8.039	2-O-CH ₃	4-NO ₂	
8.040	2-F	6-NO ₂	•
8.041	2-Cl	6-NO ₂	•
8.042	2-CH ₃	6-NO ₂	
8.043	2-O-CH ₃	6-NO ₂	
8.044	2-F	4-NO ₂	·
8.045	2-CH ₃	$4-N(C_2H_5)_2$	
8.046	2-Cl	4-SO ₂ -CH ₃	
8.047	2-C1	4-SO-CH ₃	
8.048	2-Cl	4-S-CH ₃	
8.()49	2-Cl	6-SO ₂ -CH ₃	•
8.050	2-Cl	6-SO-CH ₃	•
8.051	2-Cl	6-S-CH ₃	
8.052	2-CH ₃	4-SO ₂ -CH ₃	· · · · · · · · · · · · · · · · · ·
8.053	2-CH ₃	4-SO-CH ₃	
8.054	2-CH ₃	4-S-CH ₃	·.
3.055	2-CH ₃	6-SO ₂ -CH ₃	•
3.056	2-CH ₃	6-SO-CH ₃	
8.057	2-CH ₃	6-S-CH ₃	

Comp.		R _n	Phys. data (°C)
8.058		2-O-CH ₃	6-SO ₂ -CH ₃
8.059		2-O-CH ₃	6-SO-CH ₃
8.060		2-O-CH ₃	6-S-CH ₃
8.061		2-O-CH ₃	4-SO ₂ -CH ₃
8.062		2-O-CH ₃	4-SO-CH ₃
8.063		2-O-CH ₃	4-S-CH ₃
8.064		2-CH ₃	$6-N(C_2H_5)_2$
8.065		2-Cl	6-N(CH ₃) ₂
8.066		2-Cl	4-N(CH ₃) ₂
8.067		2-C1	4-CO ₂ CH ₃
8.068		2-CH ₃	6-CO ₂ C ₂ H ₅
8.069		2-CH ₃	$4-CO_2C_2H_5$
8.070		2-CH ₃	4-CN
8.071		2-CH ₃	6-CN
8.072		2-Cl	4-CN
8.073		2-CI	6-CN
8.074		2-Cl	4-CO-CH ₃
8.075		2-O-CHF ₂	4-O-CHF ₂
8.076		2-CH ₃	4-O-CHF ₂
8.077		2-Cl	4-O-CF ₃
8.078		2-O-CF ₃	4-O-CH ₃
8.079		2-O-CHF ₂	4-Cl
8.080		2-O-CHF ₂	6-CH₃
8.081		2-O-CHF ₂	6-Cl
8.082	2-O-CHF ₂	4-CH ₃	6-CH ₃
8.083	2-CH ₃	$4-t-C_4H_9$	6-CH ₃
8.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇
8.085	2-CH ₃	4-O-CH ₃	6-CH ₃
8.086	2-C!	4-CF ₃	6-Cl
8.087	2-Cl	4-CF ₃	6-F
8.088	2-Cl	4-NO ₂	6-Cl
8.089	2-Cl	4-Cl	6-Cl
8.090	2-F	4-F	6-F
8.091	2-CH ₃	4-NO ₂	6-CH ₃
8.092	2-Cl	4-Cl	6-CH ₃

Comp. R _n		R_n		Phys. data (°C)	
NO.					
8.093	2-Cl	4-O-CH ₃	6-Cl		
8.094	2-Cl	4-Cl	6-O-CH ₃	•	
8.095	2-F	4-O-CH ₃	6-F		
8.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃		
8.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃		
8.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃		
8.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃		
8.100	2-CH ₃	4-C1	6-CH ₃		
8.101	2-CH ₃	4-F	6-CH ₃		
8.102	2-CH ₃	4-CH ₃	6-O-CH ₃	•	
8.103	2-F	4-Cl	5-O-i-C ₃ H ₇		
8.104	2-C1	4-Cl	5-O-CH ₃		
8.105		4-Cl	5-O-CH ₃	•••	
8.106	2-F	4-Cl	5-CO-O-CH ₃		
8.107	2-F	4-Cl	5-CO-O-C ₂ H ₅		
8.108		4-Cl	5-CO-O-CH ₃		
8.109	2-Cl	4-Cl	5-CO-O-i-C ₃ H ₇		
8.110		4 - 0		· •	
8.111		4 - 0 — CI			
8.112		4 - 0 — F			
8.113		4 - O - CF ₃		-	
8.114	2-CH ₃	4-0-			
8.115		4 - 5 -			
8.116		-4 - S - CI			

Comp. No.	R _n	Phys. data (°C)
8.117	4 - CH ₂	>
8.118	4 - CH ₂ -	C1
8.119	4 - CH ₂	→ F
8.120	4 - CH ₂	-CF ₃
8.121	4 - N - CHO	
8.122 2-		5-O-CH ₂ -CH=CH ₂
8.123 2-	F 4-Cl	5-O-CH ₂ -C≡CH

Table 9

Compunds of the formula

$$\bigcap_{N} \bigcap_{O} \bigcap_{CH_3} R_n$$

Comp. No.		R _n	Phys. data (°C)	
9.001		Н		
9.002		2-CH ₃		
9.003		4-CH ₃		
9.004		2-CH ₃	4-CH ₃	
9.005	•	2-CH ₃	6-CH ₃	
9.006		2-CH ₃	5-CH ₃	-
9.007		3-CH ₃	5-CH ₃	•
9.008		2-CH ₃	3-CH ₃	
9.009		3-CH ₃	4-CH ₃	
9.010	2-CH ₃	4-CH ₃	6-CH ₃	
9.011	2-CH ₃	4-CH ₃	5-CH ₃	
9.012		2-Cl		
9.013		4-Cl		
9.014		2-Cl	4-Cl	
9.015		2-Cl	6-Cl	
9.016		2-Cl	6-F	
9.017		2-CH ₃	4-Cl	
9.018		2-CH ₃	4-F	
9.019		2-Cl	4-CH ₃	
9.020		2-Cl	6-CH ₃	
9.021		2-F	4-F	
9.022		2-F	6-F	•

Comp. No.	R _n	Phys. data (°C)
9.023	2-CH ₃	4-O-CH ₃
9.024	2-CH ₃	6-O-CH ₃
9.025	2-Cl	4-O-CH ₃
9.026	2-C1	6-O-CH ₃
9.027	3-OCH ₃	4-OCH ₃
9.028	2-OCH ₃	5-OCH ₃
9.029	2-OCH ₃	4-OCH ₃
9.030	2-OCH ₃	6-OCH ₃
9.031	2-CF ₃	6-CF ₃
9.032	2-CF ₃	4-CF ₃
9.033	3-CF ₃	5-CF ₃
9.034	2-Cl	4-CF ₃
9.035	2-Cl	6-CF ₃
9.036	.2-NO ₂	4-NO ₂
9.037	2-Cl	4-NO ₂
9.038	2-CH ₃	4-NO ₂
9.039	2-O-CH ₃	4-NO ₂
9.040	2-F	6-NO ₂
9.041	2-Cl	6-NO ₂
9.042	2-CH ₃	6-NO ₂
9.043	2-O-CH ₃	6-NO ₂
9.044	2-F	4-NO ₂
9.045	2-CH ₃	$4-N(C_2H_5)_2$
9.046	2-Cl	4-SO ₂ -CH ₃
9.047	2-Cl	4-SO-CH ₃
9.048	2-Cl	4-S-CH ₃
9.049	2-Cl	6-SO ₂ -CH ₃
9.050	2-Cl	6-SO-CH ₃
9.051	2-Cl	6-S-CH ₃
9.052	2-CH ₃	4-SO ₂ -CH ₃
9.053	2-CH ₃	4-SO-CH ₃
9.054	2-CH ₃	4-S-CH ₃
9.055	2-CH ₃	6-SO ₂ -CH ₃
9.056	2-CH ₃	6-SO-CH ₃
0.057	2-CH ₃	6-S-CH ₃

Comp. No.	·	R _n		Phys. data (°C)
9.058		2-O-CH ₃	6-SO ₂ -CH ₃	
9.059		2-O-CH ₃	6-SO-CH ₃	
9.060		2-O-CH ₃	6-S-CH ₃	
9.061		2-O-CH ₃	4-SO ₂ -CH ₃	
9.062	•	2-O-CH ₃	4-SO-CH ₃	
9.063		2-O-CH ₃	4-S-CH ₃	
9.064		2-CH ₃	$6-N(C_2H_5)_2$	
9.065		2-C1	$6-N(CH_3)_2$	
9.066		2-Cl	$4-N(CH_3)_2$	
9.067		2-Cl	4-CO ₂ CH ₃	
9.068		2-CH ₃	$6-CO_2C_2H_5$	
9.069		2-CH ₃	4-CO ₂ C ₂ H ₅	
9.070		2-CH ₃	4-CN	
9.071		2-CH ₃	6-CN	
9.072		2-Cl	4-CN	
9.073		2-Cl	6-CN	
9.074	•	2-Cl	4-CO-CH ₃	
9.075		2-O-CHF ₂	4-O-CHF ₂	
9.076		2-CH ₃	4-O-CHF ₂	·
9.077		2-Cl	4-O-CF ₃	
9.078		2-O-CF ₃	4-O-CH ₃	
9.079		2-O-CHF ₂	4-Cl	
9.080		2-O-CHF ₂	6-CH ₃	
9.081		2-O-CHF ₂	6-Cl	
9.082	2-O-CHF ₂	4-CH ₃	6-CH ₃	
9.083	2-CH ₃	4-t-C ₄ H ₉	6-CH ₃	
9.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇	
9.085	2-CH ₃	4-O-CH ₃	6-CH ₃	
9.086	2-C!	4-CF ₃	6-Cl	
9.087	2-C1	4-CF ₃	6-F	. "
9.088	2:-Cl	4-NO ₂	6-Cl	
9.089	2-Cl	4-Cl	6-Cl	•
9.090	2-F	4-F	6-F	·
9.091	2-CH ₃	4-NO ₂	6-CH ₃	•
9.092	2-Cl	4-Cl	6-CH ₃	

Comp.		R _n	Phys. data (°C)
9.093	2-Cl	4-O-CH ₃	6-C1
9.094	2-Cl	4-Cl	6-O-CH ₃
9.095	2-F	4-O-CH ₃	6-F
9.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃
9.097	_	4-O-CH ₃	6-CH ₃
9.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃
9.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃
9.100	2-CH ₃	4-Cl	6-CH ₃
9.101	2-CH ₃	4-F	6-CH ₃
9.102	2-CH ₃	4-CH ₃	6-O-CH ₃
9.103	2-F	4-Cl	5-O-i-C ₃ H ₇
9.104	2-Cl	4-Cl	5-O-CH ₃
9.105		4-Cl	5-O-CH ₃
9.106	2-F	4-Cl	5-CO-O-CH ₃
9.107	2-F	4-Cl	5-CO-O-C ₂ H ₅
9.108		4-Cl	5-CO-O-CH ₃
9.109	2-Cl	4-Cl	5-CO-O-i-C ₃ H ₇
9.110		4 - 0	•
9.111		4 - O — CI	
9.112		4 - O - F	
9.113		4-0-CF ₃	
9.114	2-CH ₃	4-0-	
9.115		4 - 8 -	
9.116		4 - S — CI	•

Comp. No.	R_n .	Phys. data (°C)
9.117	4 - CH ₂	
9.118	4 - CH ₂	- Cl
9.119	4 - CH ₂	– F
9.120	4 - CH ₂	·CF ₃
9.121	4 - N (CHO)	
9.122 2-F	4-Cl	5-O-CH ₂ -CH=CH ₂
9.123 2-F	4-Cl	5-O-CH ₂ -C≡CH

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Table 10

Comp.		R _n		Phys. data (°C)
10.001		Н		
10.002		2-CH ₃		
10.003		4-CH ₃	•	
10.004		2-CH ₃	4-CH ₃	
10.005		2-CH ₃	6-CH ₃	
10.006		2-CH ₃	5-CH ₃	
10.007		3-CH ₃	5-CH ₃	
10.008		2-CH ₃	3-CH ₃	
10.009		3-CH ₃	4-CH ₃	
10.010	2-CH ₃	4-CH ₃	6-CH ₃	m.p. 205-207
10.011	2-CH ₃	4-CH ₃	5-CH ₃	
10.012		2-Cl		
10.013		4-Cl		
10.014		2-Cl	4-Cl	
10.015		2-Cl	6-Cl	
10.016		2-Cl	6-F	
10.017		2-CH ₃	4-Cl	
10.018		2-CH ₃	4-F	
10.019		2-Cl	4-CH ₃	
10.020		2-Cl	6-CH ₃	
10.021		2-F	4-F	
10.022		2-F	6-F	

Comp.	R_n		Phys. data (°C)
No.			
	·		
10.023	2-CH ₃	4-O-CH ₃	
10.024	2-CH ₃	6-O-CH ₃	
10.025	2-Cl	4-O-CH ₃	
10.026	2-C1	6-O-CH ₃	
10.027	3-OCH ₃	4-OCH ₃	
10.028	2-OCH ₃	5-OCH ₃	
10.029	2-OCH ₃	4-OCH ₃	
10.030	2-OCH ₃	6-OCH ₃	
10.031	2-CF ₃	6-CF ₃	
10.032	2-CF ₃	4-CF ₃	
10.033	3-CF ₃	5-CF ₃	
10.034	2-Cl	4-CF ₃	
10.035	2-Cl	6-CF ₃	
10.036	$2-NO_2$	4-NO ₂	·
10.037	2-Cl	4-NO ₂	
10:038	2-CH ₃	4-NO ₂	••
10.039	2-O-CH ₃	4-NO ₂	•
10.040	2-F	6-NO ₂	
10.041	2-Cl	$6-NO_2$	
10.042	2-CH ₃	6-NO ₂	
10.043	2-O-CH ₃	6-NO ₂	
10.044	2-F	4-NO ₂	·
10.045	2-CH ₃	$4-N(C_2H_5)_2$	•
10.046	2-CI	4-SO ₂ -CH ₃	
10.047	2-Cl	4-SO-CH ₃	
10.048	2-Cl	4-S-CH ₃	
10.049	2-Cl	6-SO ₂ -CH ₃	
0.050	2-Cl	6-SO-CH ₃	
0.051	2-Cl	6-S-CH ₃	
0.052	2-CH ₃	4-SO ₂ -CH ₃	
0.053	2-CH ₃	4-SO-CH ₃	^{7.}
0.054	2-CH ₃	4-S-CH ₃	
0.055	2-CH ₃	6-SO ₂ -CH ₃	
0.056	2-CH ₃	6-SO-CH ₃	

Comp.		R_n	Ph	ys. data (°C)
10.057		2-CH ₃	6-S-CH ₃	
10.058		2-O-CH ₃	6-SO ₂ -CH ₃	
10.059		2-O-CH ₃	6-SO-CH ₃	
10.060		2-O-CH ₃	6-S-CH ₃	
10.061		2-O-CH ₃	4-SO ₂ -CH ₃	
10.062		2-O-CH ₃	4-SO-CH ₃	
10.063		2-O-CH ₃	4-S-CH ₃	
10.064		2-CH ₃	$6-N(C_2H_5)_2$	
10.065		2-Cl	$6-N(CH_3)_2$	
10.066		2-Cl	4-N(CH ₃) ₂	
10.067		2-Cl	4-CO ₂ CH ₃	
10.068		2-CH ₃	$6-CO_2C_2H_5$	
10.069		2-CH ₃	$4-CO_2C_2H_5$	
10.070		2-CH ₃	4-CN	
10.071		2-CH ₃	6-CN	
10.072		2-Cl	4-CN	
10.073		2-Cl	6-CN	
10.074		2-Cl	4-CO-CH ₃	
10.075		2-O-CHF ₂	4-O-CHF ₂	
10.076		2-CH ₃	4-O-CHF ₂	
10.077		2-Cl	4-O-CF ₃	
10.078		2-O-CF ₃	4-O-CH ₃	
10.079		2-O-CHF ₂	4-Cl	
10.080		2-O-CHF ₂	6-CH ₃	
10.081		2-O-CHF ₂	6-Cl	
10.082	2-O-CHF ₂	4-CH ₃	6-CH ₃	
10.083	2-CH ₃	4-t-C ₄ H ₉	6-CH ₃	
10.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇	
10.085	2-CH ₃	4-O-CH ₃	6-CH ₃	
10.086	2-Cl	4-CF ₃	6-Cl	
10.087	2-Cl	4-CF ₃	6-F	
10.088	2-Cl	4-NO ₂	6-Cl	
10.089	2-Cl	4-Cl	6-Cl	
10.090	2-F	4-F	6-F	
10.091	2-CH ₃	4-NO ₂	6-CH ₃	

Phys. data (°C)

Comp. No.		R _n	
10.092	2-Cl	4-Cl	6-CH ₃
10.093	2-Cl	4-O-CH ₃	6-Cl
10.094	2-Cl	4-Cl	6-O-CH ₃
10.095	2-F	4-O-CH ₃	6-F
10.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃
10.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃
10.098	2-O-CH ₃	4-O-CH ₃	6-O-CH₃
10.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃
10.100	$2-CH_3$	4-Cl	6-CH ₃
10.101	2-CH ₃	4-F	6-CH ₃
10.102	2-CH ₃	4-CH ₃	6-O-CH ₃
10.103	2-F	4-Cl	5-O-i-C ₃ H ₇
10.104	2-Cl	4-Cl	5-O-CH ₃
10.105		4-Cl	5-O-CH ₃
10.106	2-F	4-Cl	5-CO-O-CH ₃
10.107	2-F	4-Cl	5-CO-O-C ₂ H ₅
10.108		4-Cl	5-CO-O-CH ₃
10.109	2-Cl	4-Cl 5-CO-O-i-C ₃ H ₇	
10.110		4 - 0	
10.111		4 - 0 — C1	·
10.112		4 - 0 — F	
10.113		4 - O - CF ₃	
10.114	2-CH ₃	4-0-	
10.115		4 - S	
10.116		4 - S — CI	

Comp.		R _n	Phys. data (°C)
10.117		4 - CH ₂	
10.118		4 - CH ₂ —CI	
10.119		4 - CH ₂ - F	
10.120		4 - CH ₂ - CF ₃	
10.121		4 - N-CHO	
10.122	2-F	4-Cl	5-O-CH ₂ -CH=CH ₂
10.123	2-F	4-Cl	5-O-CH ₂ -C≡CH
10.124	2-Br		-
10.125	2-CF ₃		
10.126	2-OCH ₃	a.	
10.127	2-CH ₃	4 -0 -C1	
10.128	2-CH ₃	4-0-CF ₃	
10.129	2-CH ₃	4 -0 -C1	· .
10.130	2-CH ₃	4 -0 -	6-CH ₃
10.131	2-CH ₃	4-0-CF ₃	6-CH ₃
10.132	2-CH ₃	4 -0 — CI	6-CH ₃
10.133	2-CH ₃	4 -0 — CI	6-CH ₃
		Cı	

Comp.		R_n	R _n	
No.				·
	٠			.
10.134	2-CH ₃	4-Br	6-CH ₃	-
10.135	2-CH ₃	$6-C_2H_5$		
10.136	$2-C_2H_5$	$6-C_2H_5$		
10.137	$2-CH_3$	$4-OC_2H_5$	6-CH ₃	
10.138	$2-CH_3$	4-O-i-C ₃ H ₇	6-CH ₃	•
10.139	2-CH ₃	4-O-n-C ₃ H ₇	6-CH ₃	
10.140	2-CH ₃	4-O-n-C ₁₀ H ₂₁	6-CH ₃	
10.141	2-CH ₃	4-O-n-C ₃ H ₇		
10.142	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
10.143	$2-CH_3$	4-O-(CH ₂) ₂ OCH ₃	6-CH ₃	
10.144	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
10.145	2-CH ₃	4-O-n-C ₆ H ₁₃	6-CH ₃	
10.146	2-CH ₃	4-0-		
10.147	2-CH ₃	4 -o — N	6-CH₃	
10.148	2-CH ₃	4 -0 — CF ₃		
10.149	2-CH ₃	4 -0 - CF ₃	6-CH ₃	<u>.</u> .
10.150	2-CH ₃	4-0-KP3		
10.151	2-CH ₃	4-0-CF ₃	6-CH₃	
10.152	2-CH ₃	5-0-KP3		
10.153	2-CH ₃	5 -0 — CF ₃		

Comp.		R _n		Phys. data (°C)
10.154	2-CH ₃	4 - S —	6-CH ₃	
10.155	2-CH ₃	4 - s — CI	6-CH ₃	
10.156	2-C ₂ H ₅	4 - S —	6-CH ₃	

Table 11

Comp.		R_n		Phys. data (°C)
11.001		Н		
11.002		2-CH ₃		
11.003		4-CH ₃		
11.004		2-CH ₃	4-CH ₃	
11.005		2-CH ₃	6-CH ₃	
11.006		2-CH ₃	5-CH ₃	-
11.007		3-CH ₃	5-CH ₃	
11.008		2-CH ₃	3-CH ₃	
11.009		3-CH ₃	4-CH ₃	
11.010	2-CH ₃	4-CH ₃	6-CH ₃	m.p. 195-196
11.011	2-CH ₃	4-CH ₃	5-CH ₃	
11.012		2-Cl		
11.013		4-Cl		
11.014		2-Cl	4-Cl	
11.015		2 ₌ Cl	6-Cl	
11.016		2-C1	6-F	
i1.017		2-CH ₃	4-Cl	•
11.018		2-CH ₃	4-F	
11.019		2-Cl	4-CH ₃	·
11.020		2-Cl	6-CH ₃	·
11.021		2-F	4-F	
11.022	•	2-F	6-F	

Comp.	R_n		Phys. data (°C)
11.023	2-CH ₃	4-O-CH ₃	
11.024	2-CH ₃	6-O-CH ₃	
11.025	2-Cl	4-O-CH ₃	
11.026	2-Cl	6-O-CH ₃	
11.027	3-OCH ₃	4-OCH ₃	
11.028	2-OCH ₃	5-OCH ₃	
11.029	2-OCH ₃	4-OCH ₃	
11.030	2-OCH ₃	6-OCH ₃	
11.031	2-CF ₃	6-CF ₃	
11.032	2-CF ₃	4-CF ₃	
11.033	3-CF ₃	5-CF ₃	
11.034	2-Cl	4-CF ₃	
11.035	2-Cl	6-CF ₃	
11.036	$2-NO_2$	4-NO ₂	•
11.037	2-Cl	4-NO ₂	
11.038	2-CH ₃	4-NO ₂	
11.039	2-O-CH ₃	4-NO ₂	
11.040	2-F	6-NO ₂	
11.041	2-Cl	6-NO ₂	
11.042	2-CH ₃	6-NO ₂	
11.043	2-O-CH ₃	6-NO ₂	
11.044	2-F	4-NO ₂	
11.045	2-CH ₃	$4-N(C_2H_5)_2$	
11.046	2-C1	4-SO ₂ -CH ₃	
11.047	2-Cl	4-SO-CH ₃	·
11.048	2-Cl	4-S-CH ₃	
11.049	2-Cl	6-SO ₂ -CH ₃	
11.050	2-Cl	6-SO-CH ₃	
11.051	2-Cl	6-S-CH ₃	
11.052	2-CH ₃	4-SO ₂ -CH ₃	
11.053	2-CH ₃	4-SO-CH ₃	
11.054	2-CH ₃	4-S-CH ₃	•
11.055	2-CH ₃	6-SO ₂ -CH ₃	
11.056	2-CH ₃	6-SO-CH ₃	
11.057	2-CH ₃	6-S-CH ₃	

Comp.		R _n		Phys. data (°C)
11.058		2-O-CH ₃	6-SO ₂ -CH ₃	
11.059		2-O-CH ₃	6-SO-CH ₃	
11.060		2-O-CH ₃	6-S-CH ₃	
11.061		2-O-CH ₃	4-SO ₂ -CH ₃	
11.062	•	2-O-CH ₃	4-SO-CH ₃	
11.063		2-O-CH ₃	4-S-CH ₃	
11.064		2-CH ₃	$6-N(C_2H_5)_2$	
11.065		2-Cl	6-N(CH ₃) ₂	
11.066		2-Cl	4-N(CH ₃) ₂	
11.067		2-C1	4-CO ₂ CH ₃	
11.068		2-CH ₃	6-CO ₂ C ₂ H ₅	
11.069		2-CH ₃	$4-CO_2C_2H_5$	
11.070		2-CH ₃	4-CN	
11.071		2-CH ₃	6-CN	·
11.072		2-C1	4-CN	
11.073	•	2-Cl	6-CN	
11.074		2-C1	4-CO-CH ₃	
11.075		2-O-CHF ₂	4-O-CHF ₂	
11.076		2-CH ₃	4-O-CHF ₂	
11.077		2-Cl	4-O-CF ₃	
11.078		2-O-CF ₃	4-O-CH ₃	
11.079		2-O-CHF ₂	4-Cl	
11.080		2-O-CHF ₂	6-CH ₃	•
11.081		2-O-CHF ₂	6-Cl	
11.082	2-O-CHF ₂	4-CH ₃	6-CH ₃	
11.083	2-CH ₃	$4-t-C_4H_9$	6-CH ₃	
11.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇	
11.085	2-CH ₃	4-O-CH ₃	6-CH ₃	
11.086	2-C!	4-CF ₃	6-Cl	
11.087	2-Cl	4-CF ₃	6-F	
11.088	2-Cl	4-NO ₂	6-Cl	
11.089	2-C1	4-Cl	6-C1	
11.090	2-F	4-F	6-F	
11.091	2-CH ₃	4-NO ₂	6-CH ₃	
11.092	2-Cl	4-Cl	6-CH ₃	

Comp.		R _n		Phys. data (°C)
11.093	2-Cl	4-O-CH ₃	6-Cl	
11.094	2-Cl	4-Cl	6-O-CH ₃	
11.095	2-F	4-O-CH ₃	6-F	
11.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃	
11.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃	
11.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃	
11.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃	
11.100	2-CH ₃	4-Cl	6-CH ₃	
11.101	2-CH ₃	4-F	6-CH ₃	
11.102	2-CH ₃	4-CH ₃	6-O-CH ₃	
11.103	2-F	·4-Cl	5-O-i-C ₃ H ₇	
11.104	2-Cl	4-Cl	5-O-CH ₃	
11.105		4-Cl	5-O-CH ₃	
11.106	2-F	4-Cl	5-CO-O-CH ₃	
11.107	2-F	4-Cl	5-CO-O-C ₂ H ₅	·
11.108		4-Cl	5-CO-O-CH ₃	
11.109	2-Cl	4-Cl	5-CO-O-i-C ₃ H ₇	
11.110		4 - 0		
11.111		4 - 0 — CI		
11.112		4 - 0 — F		
11.113		4 - O - CF ₃		
11.114	2-CH ₃	4-0-		
11.115		4 - S —		
11.116		4 - S — CI		

Comp. No.		R _n		Phys. data (°C)
11.117		4 - CH ₂		
11.118		4 - CH ₂ —CI		
11.119		4 - CH ₂ - F		
11.120		4 - CH ₂ - CF ₃		
11.121		4 - N (CHO)		
11.122	2-F	4-Cl	5-O-CH ₂ -CH=CH ₂	
11.123	2-F	4-Cl	5-O-CH ₂ -C≡CH	
11.124	2-Br			
11.125	2-CF ₃			
11.126	2-OCH ₃		•	
11.127	2-CH ₃	4 -0 -CI		
11.128	2-CH ₃	4 - O — CF ₃		
11.129	2-CH ₃	4 -0 -C1		
11.130	2-CH ₃	4 -0 -	6-CH ₃	
11.131	2-CH ₃	4-0-CF3	6-CH ₃	
11.132	2-CH ₃	4 -0 -C1	6-Cl·l ₃	
11.133	2-CH ₃	4 -0 -CI	6-CH ₃	
		Cı		

Comp.		R_n	Phys. data (°C)	
11.134	2-CH ₃	4-Br	6-CH₃	
11.135	2-CH ₃	6-C ₂ H ₅		
11.136	$2-C_2H_5$	6-C ₂ H ₅		
11.137	2-CH ₃	4-OC ₂ H ₅	6-CH ₃	
11.138	2-CH ₃	4-O-i-C ₃ H ₇	6-CH ₃	
11.139	2-CH ₃	4-O-n-C ₃ H ₇	6-CH ₃	
11.140	2-CH ₃	4-O-n-C ₁₀ H ₂₁	6-CH ₃	
11.141	2-CH ₃	4-O-n-C ₃ H ₇		
11.142	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
11.143	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃	6-CH ₃	
11.144	2-CH ₃	4-O-(CH ₂) ₂ OCH ₃		
11.145	2-CH ₃	4-O-n-C ₆ H ₁₃	6-CH ₃	•
11.146	2-CH ₃	4-0-		
11.147	2-CH ₃	4 -o — N	6-CH ₃	
11.148	2-CH ₃	4 -0 - CF ₃		
11.149	2-CH ₃	4 -0 - CF ₃	6-CH ₃	
11.150	2-CH ₃	$4-0 \xrightarrow{N} CF_3$		
11.151	2-CH ₃	4-0-KP3	6-CH ₃	
11.152	2-CH ₃	5 - 0 — CF ₃		
11.153	2-CH ₃	5 -O - CF ₃	·	

Comp. No.		R _n		Phys. data (°C)
11.154	2-CH ₃	4 -s —	6-CH ₃	
11.155	2-CH ₃	4 - s — C1	6-CH ₃	
11.156	2-C ₂ H ₅	4 - s —	6-CH ₃	

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Table 12

Compounds of the formula

$$CH_3 \underset{CH_2}{\overset{C}{\bigvee}} \underset{CH_3}{\overset{C}{\bigvee}} \underset{O}{\overset{R_n}{\bigvee}}$$

Comp. No.	R _n	Phys. data (°C)
12.001	Н	
12.002	2-CH ₃	
12.003	4-CH ₃	
12.004	2-CH ₃	4-CH ₃
12.005	2-CH ₃	6-CH ₃
12.006	2-CH ₃	5-CH ₃
12.007	3-CH ₃	5-CH ₃
12.008	2-CH ₃	3-CH ₃
12.009	3-CH ₃	4-CH ₃
12.010 2-CH ₃	4-CH ₃	6-CH ₃
12.011 2-CH ₃	4-CH ₃	5-CH ₃
12.012	2-Cl	
12.013	4-Cl	·
12.014	2-Cl	4-Cl
12.015	2-Cl	6-Cl
12.016	2-Cl	6-F
12.017	2-CH ₃	4-Cl
12.018	2-CH ₃	4-F
12.019	2-Cl	4-CH ₃
12.020	2-Cl	6-CH ₃
12.021	2-F	4-F
12.022	2-F	6-F

Comp. No.	R _n		Phys. data (°C)
12.023	2-CH ₃	4-O-CH ₃	
12.024	2-CH ₃	6-O-CH ₃	
12.025	2-Cl	4-O-CH ₃	
12.026	2-Cl	6-O-CH ₃	
12.027	3-OCH ₃	4-OCH ₃	
12.028	2-OCH ₃	5-OCH ₃	
12.029	2-OCH ₃	4-OCH ₃	
12.030	2-OCH ₃	6-OCH ₃	
12.031	2-CF ₃	6-CF ₃	
12.032	2-CF ₃	4-CF ₃	
12.033	3-CF ₃	5-CF ₃	
12.034	2-C1	4-CF ₃	
12.035	2-Cl	6-CF ₃	
12.036	2-NO ₂	4-NO ₂	
12.037	2-Cl	4-NO ₂	
12.038	2-CH ₃	4-NO ₂	
12.039	2-O-CH ₃	4-NO ₂	
12.040	2-F	6-NO ₂	
12.041	2-C1	6-NO ₂	
12.042	2-CH ₃	6-NO ₂	•
12.043	2-O-CH ₃	6-NO ₂	
12.044	2-F	4-NO ₂	
12.045	2-CH ₃	$4-N(C_2H_5)_2$	
12.046	2-C1	4-SO ₂ -CH ₃	
12.047	2-C1	4-SO-CH ₃	
12.048	2-C1	4-S-CH ₃	
12.049	2-Cl	6-SO ₂ -CH ₃	•
12.050	2-Cl	6-SO-CH ₃	· ·
12.051	2-Cl	G-S-CH ₃	
12.052	2-CH ₃	4-SO ₂ -CH ₃	
12.053	2-CH ₃	4-SO-CH ₃	•
12.054	2-CH ₃	4-S-CH ₃	
12.055	2-CH ₃	6-SO ₂ -CH ₃	
12.056	2-CH ₃	6-SO-CH ₃	
12.057	2-CH ₃	6-S-CH ₃	

Comp.		R_n	Phys. data (°C)
No.			
12.058		2-O-CH ₃	6-SO ₂ -CH ₃
12.059		2-O-CH ₃	6-SO-CH ₃
12.060		2-O-CH ₃	6-S-CH₃
12.061		2-O-CH ₃	4-SO ₂ -CH ₃
12.062		2-O-CH ₃	4-SO-CH ₃
12.063		2-O-CH ₃	4-S-CH ₃
12.064		2-CH ₃	$6-N(C_2H_5)_2$
12.065		2-Cl	6-N(CH ₃) ₂
12.066		2-Cl	4-N(CH ₃) ₂
12.067		2-Cl	4-CO ₂ CH ₃
12.068		2-CH ₃	6-CO ₂ C ₂ H ₅
12.069		2-CH ₃	4-CO ₂ C ₂ H ₅
12.070		2-CH ₃	4-CN
12.071		2-CH ₃	6-CN
12.072		2-Cl	4-CN
12.073	•	2-Cl	6-CN
12.074		2-Cl	4-CO-CH ₃
12.075		2-O-CHF ₂	4-O-CHF ₂
12.076		2-CH ₃	4-O-CHF ₂
12.077		2-Cl	4-O-CF ₃
12.078		2-O-CF ₃	4-O-CH ₃
12.079		2-O-CHF ₂	4-Cl
12.080		2-O-CHF ₂	6-CH ₃
12.081		2-O-CHF ₂	6-Cl
12.082	2-O-CHF ₂	4-CH ₃	6-CH₃
12.083	2-CH ₃	4-t-C ₄ H ₉	6-CH ₃
12.084	2-i-C ₃ H ₇	4-i-C ₃ H ₇	6-i-C ₃ H ₇
12.085	2-CH ₃	4-O-CH ₃	6-CH ₃
12.086	2-Cl	4-CF ₃	6-Cl
12.087	2-CI	4-CF ₃	6-F
12.088	2-Cl	4-NO ₂	6-Cl
12.089	2-Cl	4-Cl	6-Cl
12.090	2-F	4-F	6-F
12.091	2-CH ₃	4-NO ₂	6-CH₃
12.092	2-Cl	4-Cl	6-CH ₃

Comp. No.		R _n		Phys. data (°C)
12.093	2-Cl	4-O-CH ₃	6-Cl	
12.094	2-Cl	4-Cl	6-O-CH ₃	
12.095	2-F	4-O-CH ₃	6-F	·
12.096	2-O-CH ₃	4-CH ₃	6-O-CH ₃	·
12.097	2-O-CH ₃	4-O-CH ₃	6-CH ₃	
12.098	2-O-CH ₃	4-O-CH ₃	6-O-CH ₃	
12.099	2-O-CH ₃	4-CO-O-CH ₃	6-O-CH ₃	
12.100	2-CH ₃	4-Cl	6-CH ₃	
12.101	2-CH ₃	4-F	6-CH ₃	
12.102	2-CH ₃	4-CH ₃	6-O-CH ₃	·
12.103	2-F	4-Cl	5-O-i-C ₃ H ₇	
12.104	2-Cl	4-Cl	5-O-CH ₃	
12.105		4-Cl	5-O-CH ₃	
12.106	2-F	4-Cl	5-CO-O-CH ₃	
12.107	2-F	4-Cl	5-CO-O-C ₂ H ₅	
12.108		4-Cl	5-CO-O-CH ₃	
12.109	2-C1	4-Cl	5-CO-O-i-C ₃ H ₇	
12.110		4 - 0		
12.111		4 - 0 - CI		
12.112		4 - 0 — F		
12.113	٠	4 - O - CF ₃		
12.114	2-CH ₃	4-0-		
12.115		4 - S —		
- 12.116	·	4 - S — CI		

Comp. No.	R _n	Phys. data (°C)
12.117	4 - CH ₂	
12.118	4 - CH ₂ —C	
12.119	4 - CH ₂ — F	
12.120	4 - CH ₂ - CF	-3
12.121	4 - N-(CHO)	
12.122 2-F	4-Cl	5-O-CH ₂ -CH=CH ₂
12.123 2-F	4-Cl	5-O-CH ₂ -C≡CH

Table 13

Comp. No.	R _n	Phys. data (°C)
13.001	Н	m.p. 171-172
13.002	2-CH ₃	
13.003	2-OCH ₃	
13.004	4-Cl	
13.005	4-F	
13.006	4-NO ₂	
13.007	6-NO ₂	
13.008	7-NO ₂	

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Table 14

Comp. No.	R _n	Phys. data (°C)
14.001	Н	
14.002	2-CH ₃	
14.003	2-OCH ₃	
14.004	4-Cl	
14.005	4-F	
14.006	4-NO ₂	
14.007	6-NO ₂	
14.008	7-NO ₂	

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Formulation examples

Example F 1: Formulation examples of active ingredients of the formula I (% = per cent by weight)

a) Wettable powder	a)		b) c)		c)		
Active ingredient No. 1.010	20	.%	50	%	0,5	%	
Sodium ligninsulfonate	5	%	5	%	4	%	
Sodium lauryl sulfate	3	%	-	%	-	%	
Sodium diisobutylnaphthalene							
sulfonate	-		6	. %	6	%	
Octylphenol polyethylene glycol							
ether(7-8 mol of EO)	-		2	%	2	%	
Highly-disperse silica	5	%	27	%	27	%	
Kaolin	67	%	10	%	-		
Sodium chloride	-		-		59.5	%	

The active ingredient is mixed thoroughly with the additives, and the mixture is ground thoroughly in a suitable mill. This gives wettable powders which can be diluted with water to give suspensions of any desired concentration.

b) Emulsion concentrate	a)		b)	
Active ingredient No. 1.010	10	%	1	%
Octylphenol polyethylene glycol				
ether (4-5 mol of EO)	3	%	3	%
Calcium dodecylbenzene sulfonate	3	%	3	%
Castor oil polyglycol ether				
(36 mol EO)	4	%	4	%
Cyclohexanone	30	%	10	%
Xylene mixture	50	%	79	%

Emulsions of any desired concentration can be prepared from this concentrate by dilution with water.

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c) <u>Dusts</u>	a)		b)	
Active ingredient No. 1.010	0.1	%	1	%
Talc	99.9	%	-	
Kaolin	-		99	%

Ready-for-use dusts are obtained by mixing the active ingredient with the carrier and grinding the mixture on a suitable mill.

d) Extruder granules	a)		b)	
Active ingredient No. 1.010	10	%	1	%
Sodium ligninsulfonate	2	%	2	%
Carboxymethylcellulose	1	%	1	%
Kaolin	87	%	96	%

The active ingredient is mixed with the additives, and the mixture is ground and moistened with water. This mixture is extruded and subsequently dried in a stream of air.

e) Coated granules

Active ingredient No. 1.010			3	%
Polyethylene glycol (MW 200)	3	%		
Kaolin	94	%		

In a mixer, the kaolin which has been moistened with polyethylene glycol is uniformly coated with the finely-ground active ingredient. In this manner, non-dusty coated granules are obtained.

f) Suspension concentrate	a)		b)	
Active ingredient No. 1.010	40	%	5	%
Ethylene glycol	10	%	10	%
Nonylphenol polyethylene glycol				
ether (15 mol EO)	6	%	1	%
Sodium ligninsulfonate	10	%	5	%
Carboxymethylcellulose	1	%	1	%
37 % aqeuous formaldehyde				
solution	0.2	%	0.2	%
Silicone oil in the form of				

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a 75 % aqueous emulsion	0.8	%	0.8	%
Water	32	%	77	%

The finely-ground active ingredient is mixed intimately with the additives. In this manner, a suspension concentrate is obtained from which suspensions of any desired concentration can be prepared by dilution with water.

g) Salt solution Active ingredient No. 1.010 Isopropylamine Octylphenol polyethylene glycol ether (78 mol EO) Water 91 5 % 91

Biological examples

Example B 1: Pre-emergence herbicidal action

In the greenhouse, the test plants are sown in pots, and the soil surface is then immediately treated with an aqueous dispersion of the active ingredients, obtained from a 25 % emulsion concentrate, at a rate of application of 4 kgAS/ha. The pots are kept in the greenhouse at 22-25°C and 50-70 % relative atmospheric humidity, and the test is evaluated after 3 weeks.

The herbicidal action is evaluated by comparison with the untreated control group, using a 9-step rating key (1 = total damage of the test plant, 9 = no herbicidal action on the test plant).

Rating figures of 1 to 4 (in particular 1 to 3) suggest a good to very good herbicidal action.

In this test, the compounds of Tables 1 to 14 show good herbicidal action. The results for compound No. 1.010 are compiled in Table 15:

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Table 15: Pre-emergence herbicidal action

Comp.		Test plan	Test plants				
No.	Avena	Sinapis	Sctaria	Stellaria			
1.010	1	1	1	6	-		

Example B2 Post-emergence herbicidal action

A number of weeds, both monocotyledon and dicotyledon, are sprayed after emergence (in the 4- to 6-leaf stage) with an aqueous dispersion of the active ingredient at a dosage rate of 4 kgAS/ha, and the plants are maintained at 24 to 26°C and 40 to 60 % relative atmospheric humidity. The test is evaluated 15 days after the treatment.

The herbicidal action is rated analogously to Example B1.

In this test, the compounds of Tables 1 to 14 show good herbicidal action. The results for compound No. 1.010 are compiled in Table 16:

Table 16: Post-emergence herbicidal action

		TEST PLANT				Phaseolus	
	Avena	Setaria	Lolium	Solanum	Sinapis	Stellaria	vulg.
Comp. No. 1.010	1	1	1	2	2	2	2

B3 Herbicidal action against weeds in paddy rice

The aquatic weeds are sown in plastic beakers (surface area 60 cm², volume 500 ml). After sowing, the beakers are filled with water up to the soil surface. 3 days after sowing, the water level is increased to just above the soil surface (3-5 mm). Application is effected 3 days after sowing by spraying the containers with an aqueous dispersion of the test

substance. The dosage rate corresponds to a rate of application of 4 kgAS/ha (amount of spray mixture: approx. 550 l/ha).

The beakers with the plants are placed in a greenhouse under optimum growth conditions for the rice weeds (at 25 - 30°C and high atmospheric humidity).

Depending on the growth rate and plant species, the tests are evaluated 2-3 weeks after application. Rating is effected analogously to the rating key mentioned in Example 1.

In this test, the compounds of Tables 1 to 14 show good herbicidal action. The results of compound No. 1.010 are compiled in Table 17:

Table 17 Herbicidal action for paddy rice

Comp.	Tes		
No.	Echinochloa	Monochoria	
			
1.010	1	1	

B4 Action against Nilaparvata lugens

Rice plants are treated with an aqueous emulsion spray mixture containing 400 ppm of the active ingredient. After the spray coating has dried on, the rice plants are populated with cicada larvae of stage 2 and 3. The test is evaluated after 21 days. The percentage reduction of the population (% action) is determined by comparing the number of surviving cicadas on the treated with those on the untreated plants.

In this test, the compounds of Tables 1 to 14 show a good action against Nilaparvata lugens. In particular Compounds 1.010 and 1.015 show an action of more than 80 %.

B5 Action against Nephotettix cincticeps

Rice plants are treated with an aqueous emulsion spray mixture containing 400 ppm of the active ingredient. After the spray coating has dried on, the rice plants are populated with cicada larvae of stage 2 and 3. The test is evaluated after 21 days. The percentage reduction of the population (% action) is determined by comparing the number of surviving cicadas on the treated with those on the untreated plants.

In this test, compounds of Tables 1 to 14 show a good action against Nephotettix cincticeps. In particular compound 1.010 shows an action of more than 80 %.

B6 Action against Bemisia tabaci

Dwarf bean plants are placed in gauze cages and populated with Bemisia tabaci adults (whitefly). After oviposition, all adults are removed, and, after 10 days, the plants together with the nymphs are treated with an aqueous emulsion spray mixture of the active ingredients to be tested (concentration 400 ppm). The test is evaluated 14 days after application of the active ingredient by calculating the percentage hatching rate compared with the untreated control batches.

In this test, compounds according to Tables 1 to 14 show a good action against Bemisia tabaci. In particular, compounds 1.010, 1.015 and 1.086 show an action of more than 80 %.

B7 Action against Tetranychus urticae

Young bean plants are populated with a mixed population of Tetranychus urticae and, after 1 day, sprayed with an aqueous emulsion spray mixture containing 400 ppm of the active ingredient. The plants are subsequently incubated for 6 days at 25°C and then evaluated. The percentage reduction of the population (% action) is determined by comparing the number of dead eggs, larvae and adults on the treated plants with those on the untreated plants.

In this test, compounds of Tables 1 to 14 show a good action against Tetranychus urticae. In particular, compounds 1.002, 1.003, 1.004, 1.005, 1.006, 1.010, 1.012, 1.014, 1.015, 1.016, 1.086, 1.125, 2.008, 3.010, 3.015, 6.010, 10.010 and 14.001 show an action of more than 80 %.

B8 Action against Tetranychus urticae

Young bean plants are populated with a number of female Tetranychus urticae, which are removed after 24 hours. The plants which are populated with eggs are sprayed with an aqueous emulsion spray mixture containing 400 ppm of the active ingredient. The plants are subsequently incubated for 6 days at 25°C and then evaluated. The percentage reduction of the population (% action) is determined by comparing the number of dead eggs, larvae and adults on the treated plants with those on the untreated plants.

In this test, compounds of Tables 1 to 14 show a good action against Tetranychus urticae. In particular, compounds 1.002, 1.003, 1.004, 1.005, 1.006, 1.010, 1.012, 1.014, 1.015, 1.016, 1.086, 1.125, 2.008, 3.010, 3.015, 6.010, 10.010 and 14.001 show an action of more than 80 %.

B9 Action against Panonychus ulmi (OP- and carb-resistant)

Apple seedlings are populated with a number of adult female Panonychus ulmi. After seven days, the infected plants are sprayed to drip point with an aqueous emulsion spray mixture containing 400 ppm of the test compound and grown in the greenhouse. The test is evaluated 14 days later. The percentage reduction of the population (% action) is determined by comparing the number of dead spider mites on the treated plants with those on the untreated plants.

In the above test, compounds of Tables 1 to 14 show a good action. In particular, compounds 1.002, 1.004, 1.010, 1.012, 1.014, 1.015, 1.016 and 1.086 show an action of above 80 %.

WHAT IS CLAIMED IS:

1. A pyrazolidine-3,5-dione of the formula I

$$R_2$$
 N R_1 R_3 N R_1 R_1 R_1 R_2 R_3 R_1 R_2 R_3 R_1 R_3 R_1 R_3 R_3 R_1 R_3 $R_$

in which

$$R_1$$
 is $(R_4)_n$; $(R_5)_m$; or $(R_5)_m$

 R_2 and R_3 independently of one another are C_1 - C_6 alkyl; C_3 - C_6 alkenyl; or C_3 - C_6 alkynyl; or R_2 and R_3 together are a -(CH₂)₃-, -(CH₂)₄-, -CH₂-CH=CH-CH₂-, -CH₂-CH=CH- or -(CH₂)₂-CH=CH- bridge which is unsubstituted or up to trisubstituted by C_1 - C_4 alkyl;

n is 0; 1; 2; 3; or 4;

m is 0; or 1; the total of m and n being less than, or equal to, 4; the

R₄ radicals independently of one another are halogen; nitro; cyano, C₁-C₄alkyl; C₁-C₄haloalkyl; C₁-C₄haloalkoxy; C₁-C₄haloalkoxy; C₃-C₆alkenyloxy; C₁-C₄alkoxy-C₂-C₄alkoxy; C₃-C₆alkynyloxy; C₁-C₄alkylcarbonyl; C₁-C₄alkoxycarbonyl; C₁-C₄alkylthio; C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino; mono-C₁-C₄alkylamino; di-C₁-C₄alkylamino;

$$R_5 = is - x - (R_6)_0$$
; $-x - (R_6)_0$

X is oxygen: sulfur; CH₂; or NR₇;

o is 0; 1; 2; or 3;

R₆ radicals independently of one another are C₁-C₄alkyl; halogen; C₁-C₄haloalkyl; C₁-C₄haloalkoxy; C₁-C₄alkoxy; nitro; cyano; C₁-C₄alkoxycarbonyl; amino; mono-C₁-C₄alkylamino; or di-C₁-C₄alkylamino; and

 R_7 is hydrogen; C_1 - C_4 alkyl; formyl; or C_1 - C_4 alkylcarbonyl, or a acid addition salt thereof.

2. A pyrazolidine-3,5-dione according to claim 1, of the formula I in which

$$\begin{array}{c|c}
R_2 & O \\
I & \\
R_3 & N
\end{array}$$
(I),

$$R_1$$
 is $(R_4)_n$; $(R_4)_n$; or $(R_4)_n$; $(R_5)_m$; or

n is 0; 1; 2; 3; or 4;

m is 0; or 1; and the total of m and n is less than, or equal to, 4;

n' is 0; 1; 2; or 3;

n" is 0; 1; or 2;

m' is 0; or 1; and the total of m' and n' is less than, or equal to, 3; and the radicals R_1 to R_5 are as defined above.

3. A pyrazolidine-3,5-dione according to claim, of the formula I

$$R_2$$
 R_3
 R_1
 R_3
 R_1
 R_1
 R_1
 R_1
 R_2
 R_1
 R_3

in which

$$R_1$$
 is $(R_4)_n$; or $(R_5)_m$;

 R_2 and R_3 independently of one another are C_1 - C_6 alkyl; C_3 - C_6 alkenyl; or C_3 - C_6 alkynyl; or R_2 and R_3 together are a -(CH₂)₃-, -(CH₂)₄-, -CH₂-CH=CH-CH₂-, -CH₂-CH=CH- or -(CH₂)₂-CH=CH- bridge which is unsubstituted or up to trisubstituted by C_1 - C_4 alkyl;

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is 0; 1; 2; 3; or 4; n

is 0; or 1; the total of m and n being less than, or equal to, 4; m

radicals independently of one another are halogen; nitro; cyano; C₁-C₄alkyl; R_4 C₁-C₄haloalkyl; C₁-C₄alkoxy; C₁-C₄haloalkoxy; C₃-C₆alkenyloxy; $C_3\text{-}C_6 alkynyloxy; \ C_1\text{-}C_4 alkylcarbonyl; \ C_1\text{-}C_4 alkoxycarbonyl; \ C_1\text{-}C_4 alkylthio; \\$ C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino; mono-C₁-C₄alkylamino; di-C₁-C₄alkylamino;

$$R_5$$
 is $-x$ $(R_6)_0$;

is oxygen; sulfur; CH2; or NR7;

is 0; 1; 2; or 3; 0

radicals independently of one another are C₁-C₄alkyl; halogen; R_6 C₁-C₄haloalkyl;C₁-C₄haloalkoxy; C₁-C₄alkoxy; nitro; cyano; C₁-C₄alkoxycarbonyl; amino; mono-C₁-C₄alkylamino; or di-C₁-C₄alkylamino; and is hydrogen; C₁-C₄alkyl; formyl; or C₁-C₄alkylcarbonyl R_7

4. A pyrazolidine-3,5-dione according to claim 3, of the formula I

$$\begin{array}{c}
R_2 \\
N \\
I \\
R_3
\end{array}$$
 R_1 (I),

in which

$$R_1$$
 is $(R_4)_n$ or $(R_5)_m$;

is 0; 1; 2; 3; or 4; n

is 0; or 1; and the total of m and n is less than, or equal to, 4; m

n' is 0: 1: 2: or 3:

is 0; or 1; and the total of m' and n' is less than, or equal to, 3; and the radicals R_2 to R_5 are as defined above.

5. A 5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)- dione according to claim 3,

of the formula Ia

in which

 R_8 is C_1 - C_4 alkyl; and

p is 0, 1, 2 or 3, preferably 0.

6. A 5,8-dihydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione, according to claim 3, of the formula Ib

$$(R_8)_p$$
 R_1
 $(Ib)_n$

in which

 R_8 is C_1 - C_4 alkyl; and

p is 0, 1, or 2, preferably 0.

7. A 7.8-dihydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione, according to claim 3, of the formula Ic

$$(R_8)_{\overline{p}} \xrightarrow{N} \stackrel{O}{\underset{N}{\bigvee}} R_1 \text{ (Ic)}$$

in which

 R_8 is C_1 - C_4 alkyl; and

p is 0, 1, 2 or 3, preferably 0.

8. A 6,7-dihydro-1H,5H-pyrazolo[1,2-a]pyrazole-1,3(2H)-dione, according to claim 3, of the formula Id

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$$(R_8)_p$$
 N
 R_1 (Id),

in which

R₈ is C₁-C₄alkyl; and

p is 0, 1, 2 or 3, preferably 0.

9. A 1H,5H-pyrazolo[1,2-a]pyrazole-1,3(2H)-dione, according to claim 3, of the formula Ie

$$(R_g)_p$$
 N
 R_1 (Ie),

in which

 R_8 is C_1 - C_4 alkyl; and

p is 0, 1, 2 or 3, preferably 0.

10. A pyrazolidine-1,3-dione, according to claim 3, of the formula If

$$R_2$$
 N R_3 R_1 (If)

in which

R₂ and R₃ independently of one another are C₁-C₆alkyl; C₃-C₆alkenyl; or C₃-C₆alkynyl.

11. A pyrazolidine-1,3-dione, according to claim 3, of the formula Ig

in which

R₂ and R₃ independently of one another are C₁-C₆alkyl; or C₃-C₆alkenyl.

12. A pyrazolidine-1,3-dione, according to claim 3, of the formula Ih

$$R_2$$
 R_3
 R_1
 R_3
 R_1
 R_1
 R_1
 R_1
 R_1

in which

R₂ and R₃ are C₁-C₆alkyl.

13. A compound according to one of claims 5 to 12, in which

$$R_1$$
 is $N = \begin{pmatrix} (R_4)_n \\ (R_5)_m \end{pmatrix}$ and

n' is 0; 1; 2; or 3;

m' is 0; or 1; and the total of m' and n' is less than, or equal to, 3;

R₄ is not more than three times halogen; or C₁-C₄alkyl;
not more than twice C₁-C₄alkoxy; C₁-C₄haloalkoxy; C₁-C₄alkylthio;
C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino; mono-C₁-C₄alkylamino;
di-C₁-C₄alkylamino; or C₁-C₄haloalkyl; and
not more than once nitro; cyano; C₁-C₄alkylcarbonyl; C₁-C₄alkoxycarbonyl; or

$$R_1$$
 is $(R_4)_m$

n is 0; 1; 2; 3; or 4;

m is 0; or 1; and the total of m and n is less than, or equal to, 4;

Is not more than four times halogen; or C_1 - C_4 alkyl; not more than three times C_1 - C_4 alkoxy; C_1 - C_4 haloalkoxy; or C_1 - C_4 alkylthio; and not more than twice nitro; C_1 - C_4 alkylsulfinyl; C_1 - C_4 alkylsulfonyl; amino; mono- C_1 - C_4 alkylamino; di- C_1 - C_4 alkylamino; C_1 - C_4 haloalkyl; or cyano; not more than once C_1 - C_4 alkylcarbonyl; C_3 - C_6 alkenyloxy; C_3 - C_6 alkynyloxy; C_1 - C_4 alkoxycarbonyl; and

R₅ is as defined under claim 1 or 2, and the meaning of the substituent R₄ can in each case be identical or different.

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14. A compound as claimed in one or more of claims 3 to 13, in which

R₁ is
$$(R_4)_n$$
, and

n' is 0; 1; 2; or 3;

m' is 0;

R₄ is not more than three times halogen; or C₁-C₄alkyl;

not more than twice C₁-C₄alkoxy; C₁-C₄haloalkyl; C₁-C₄haloalkoxy;

C₁-C₄alkylthio; C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino;

mono-C₁-C₄alkylamino; or di-C₁-C₄alkylamino; and

not more than once nitro; cyano; C₁-C₄alkylcarbonyl; C₁-C₄alkoxycarbonyl; or

(R₄)_n

is 0; 1; 2; or 3;

m is 0; or 1; and the total of m and n is less than, or equal to, 3;

is not more than three times fluorine; chlorine; or C₁-C₄alkyl;

not more than twice C₁-C₄alkoxy; C₁-C₄haloalkyl; C₁-C₄haloalkoxy; or

C₁-C₄alkylthio; and

not more than once nitro; C₁-C₄alkylsulfinyl; C₁-C₄alkylsulfonyl; amino;

mono-C₁-C₄alkylamino; di-C₁-C₄alkylsulfinyl; C₁-C₄alkylcarbonyl;

C₃-C₆alkenyloxy; C₃-C₆alkynyloxy; or C₁-C₄alkoxycarbonyl; and the meaning of

15. A compound as claimed in one or more of claims 3 to 14, in which

the substituent R₄ can in each case be identical or different.

R₁ is
$$(R_4)_n$$
, and $(R_5)_m$, and $(R_5)_m$, is 0; 1; 2; or 3; m' is 0;

R₄ is not more than three times fluorine; chlorine; or C₁-C₂alkyl; not more than twice C₁-C₂alkoxy; C₁-C₂haloalkyl; C₁-C₂haloalkoxy; C₁-C₂alkylthio; C₁-C₂alkylsulfinyl; C₁-C₂alkylsulfonyl; amino;

mono- C_1 - C_2 alkylamino; or di- C_1 - C_2 alkylamino; and not more than once nitro; cyano; C_1 - C_2 alkylcarbonyl; C_1 - C_2 alkoxycarbonyl;

or

$$R_1$$
 is $(R_4)_n$

n is 0; 1; 2; or 3;

m is 0; or 1; and the total of m and n is less than, or equal to, 3;

is not more than three times fluorine; chlorine; or C_1 - C_4 alkyl; not more than twice C_1 - C_2 alkoxy; C_1 - C_2 haloalkyl; C_1 - C_2 haloalkoxy; or C_1 - C_2 alkylthio; and not more than once nitro; C_1 - C_2 alkylsulfinyl; C_1 - C_2 alkylsulfonyl; amino; mono- C_1 - C_2 alkylamino; di- C_1 - C_2 alkylamino; cyano; C_1 - C_2 alkylcarbonyl; C_1 - C_2 alkoxycarbonyl; and the meaning of the substituent R_4 can in each case be identical or different.

16. A pyrazolidine-3,5-dione according to claim 1, of the formula I

$$\begin{array}{c|c}
R_2 & O \\
I & R_1
\end{array}$$
(I)

in which R₁ can be the following groups

in which R_2 , R_3 , R_4 , R_5 , m and n are as defined in claim 1 and R_9 is halogen, C_1 - C_4 alkyl or C_1 - C_4 haloalkyl, the total of m + n being less than, or equal to, 3.

17. A pyrazolidine-3,5-dione according to claim 1, in which R_1 has the meaning given in claim 16 and R_2 is methyl and R_3 is methyl or ethyl, or R_2 and R_3 together are -(CH₂)₃-, -(CH₂)₄- or -CH₂-CH-CH-CH₂- .

CH₃CH₃

18. A pyrazolidine-3,5-dione of the formula I according to claim 1, in which R₁ is

$$R_1$$
 is R_{10} or 2-naphthyl

in which

R₉ is halogen; C₁-C₄alkyl; C₁-C₄haloalkyl;

R₁₀ is hydrogen; halogen; C₁-C₄alkyl; C₁-C₄haloalkyl and

 R_{11} is hydrogen; halogen or C_1 - C_4 alkyl.

19. A pyrazolidine-3,5-dione according to claim 17, in which R₁ is 2-naphthyl or

in which,

 R_0 is chlorine; C_1 - C_2 alkyl; C_1 - C_2 haloalkyl;

R₁₀ is hydrogen; chlorine; fluorine; C₁-C₂alkyl or C₁-C₂haloalkyl; and

R₁₁ is hydrogen; fluorine; chlorine or methyl.

2(). A compound of the formula Ia according to claim 5, selected from the group comprising

2-(phenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

2-(2-methylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

2-(4-methylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

2-(2,4,6-trimethylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

2-(4-chlorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

2-(2,4-dichlorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2 -a]pyridazine-1,3(2H)-dione and

2-(2,6-dichlorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

2-(2,4-dimethylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

2-(2-chlorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

2-(2-chloro-6-fluorophenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]pyridazine-1,3(2H)-dione,

1,2-dimethyl-4-(2,4,6-trimethylphenyl)-3,5-pyrazolidinedione, in particular 2-(2,4,6-trimethylphenyl)-5,6,7,8-tetrahydro-1H-pyrazolo[1,2-a]-pyrid azine-1,3(2H)-dione.

21. A process for the preparation of a compound of the formula I according to one of claims 1 to 19, which comprises

a) the cyclisation of a hydrazinecarboxylate of the formula II.

in which R_1 , R_2 and R_3 are as defined above and Y is C_1 - C_6 alkyl, phenyl or benzyl;

b) the condensation of a malonic acid derivative of the formula III, in which R_1 is as defined above, with a hydrazine derivative of the formula IV, in which R_2 and R_3 are as defined above,

and in which Y is OH, halogen or C₁-C₄alkoxy;

c) the reaction of a pyrazolidine-3,5-dione of the formula XXXIV

in which the radicals R_2 and R_3 are as defined above, with a compound of the formula XXXV

$$X-R_1$$
 (XXXV),

in which X and R_1 are as defined above, in the presence of a base or in the presence of Cu(I) or of a Pd catalyst.

22. A process for the preparation of a compound of the formula Ic and Ie, according to one or more of claims 7, 9 or 13 to 15, which comprises reacting an alcohol of the formula XIII

$$(R_S)_p \xrightarrow{(CH_2)_x} O \qquad \qquad (Ie)$$

$$(XIII)$$

in which x is 0 or 1, in the presence of an acid, to give Ic or Ie.

23. A process for the preparation of a pyrazolidine-1,3-dione of the formula If, according to one or more of claims 10 or 13 to 15, which comprises acylating a hydrazone of the

formula XIV in which R_2 is as defined above and the radical R''_3 C = is a

 C_1 - C_6 alkylidene, C_1 - C_6 alkenylidene or C_1 - C_6 alkynylidene radical, with a chloroformate IX in which Y is C_1 - C_4 alkyl, to give the N-acylhydrazone XV,

and subsequently acylating the N-acylhydrazone with arylacetyl halide of the formula X in which R_1 is as defined above and Z is chlorine or bromine, to give a hydrazine of the formula XVI,

and subsequently cyclising the hydrazine of the formula XVI in the presence of a base to give the pyrazolidine-1,3-dione of the formula If.

24. A compound of the formula II

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$$R_{2,N}$$
, COOY
$$R_{3}$$
, C CH₂-R₁ (II),
0

in which the radicals R_1 , R_2 and R_3 are as defined in one more of claims 1 to 16 and Y is C_1 - C_6 alkyl, phenyl or benzyl.

25. A process for the preparation of a compound of the formula II according to claim 24, which comprises the N-acylation of an N-acylhydrazine of the formula XVII, in which R_2 and R_3 are as defined above, and Y is C_1 - C_4 alkyl, with an arylacetyl halide of the formula X, in which R_1 is as defined above and Z is chlorine or bromine,

26. A compound of the formula XVI

in which R_1 is as defined in claim 1, Y is C_1 - C_4 alkyl and R_2 and R_3 independently of one another are C_1 - C_6 alkyl, C_3 - C_6 alkenyl or C_3 - C_6 alkynyl.

27. A process for the preparation of a compound of the formula (XVI) according to claim 26, which comprises acylating a hydrazone of the formula XIV in which R_2 is as defined

above and the radical R''_3 $C = is a C_1-C_6$ alkylidene, C_1-C_6 alkenylidene or

 C_1 - C_6 alkynylidene radical, with a chloroformate IX in which Y is C_1 - C_4 alkyl, to give an N-acylhydrazone XV,

and subsequently acylating the N-acylhydrazone with an arylacetal halide of the formula X in which R_1 is as defined above and Z is chlorine or bromine, to give a hydrazine of the formula XVI

28. An alcohol of the formula XIII"

in which R₁, R₈ and p are as defined in claim 1.

29. An N-acylhydrazine of the formula

in which R_2 and R_3 are as defined in claim 1 and in which Y is C_1 - C_4 alkyl.

30. A process for the preparation of a compound according to claim 29, which comprises

hydrolysing and decarboxylating a hydrazinedicarboxylate XII in which R_2 and R_3 are as defined above and Y is C_1 - C_4 alkyl

$$R_{2}$$
, COOY
$$R_{3}$$
 N

$$(XXII)$$

$$R_{2}$$

$$N$$

$$R_{3}$$

$$NH$$

$$(XVII)$$

31. A tetrahydropyridazinecarboxylate of the formula XXIII

in which R₈ and p are as defined in claim 1 and Y is C₁-C₄alkyl.

32. A process for the preparation of a compound according to claim 31, which comprises reacting a diene of the formula XXIV in which R_8 and p are as defined above, with an azodicarboxylate of the formula XXV, in which Y is C_1 - C_4 alkyl,

$$(R_8)_p$$
 + $(XXIII)$ $(XXIV)$ (XXV)

33. A compound of the formula XIV

in which the radical R''_3 C = is a C_1 - C_6 alkylidene, C_1 - C_6 alkenylidene or C_1 - C_6 alkynylidene radical, and R_2 is as defined in claim 1.

34. A compound of the formula XV

$$R''_{3}$$
 (XV),

in which the radical R''_3 C = is a C_1 - C_6 alkylidene, C_1 - C_6 alkenylidene or C_1 - C_6 alkynylidene radical and Y is C_1 - C_4 alkyl, and R_2 is as defined in claim 1.

35. A compound of the formula XVI

$$R_{2}$$
, CO-OY
$$R_{3}$$
 $\stackrel{N}{\sim}_{C}$
 $\stackrel{C}{\sim}^{CH_{2}}$
 $\stackrel{R_{1}}{\circ}$
 (XVI)

in which R_1 , R_2 and R_3 are as defined in claim 1 and Y is C_1 - C_4 alkyl.

36. A compound of the formula VII

in which R₈ and p are as defined in claim 1 and Y is C₁-C₄alkyl.

37. A compound of the formula XI

in which R₈, R₁ and p are as defined in claim 1 and Y is C₁-C₄alkyl.

38. A compund of the formula XII

$$(R_8)_p$$
 N
 N
 $R_1(XII),$

in which R₈, R₁ and p are as defined in claim 1.

39. A compound of the formula XIX

in which R_8 and p are as defined in claim 1 and Y is $C_1\text{-}C_4$ alkyl.

40. A compound of the formula XX

$$(R_8)_p \xrightarrow{O}_{N-C} CH_2 R_1 (XX),$$

in which R_8 , R_1 and p are as defined in claim 1 and Y is C_1 - C_4 alkyl.

41. A compound of the formula XXI

$$(R_8)_p \xrightarrow{O} R_1 \quad (XXI),$$

in which R_8 , R_1 and p are as defined in claim 1 and Y is C_1 - C_4 alkyl.

- 42. A herbicidal composition, which comprises one or more active ingredients of the formula I according to one of claims 1 to 20.
- 43. A method of controlling undesirable vegetation, wherein an effective amount of a

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composition comprising an active ingredient of the formula I according to one of claims 1 to 20 is applied to plants or their environment.

- 44. A method according to claim 43, for the selective pre- or post-emergence control of weeds in crops of useful plants.
- 45. An insecticidal or acaricidal composition which comprises one or more active ingredients of the formula I according to one of claims 1 to 20.
- 46. A method of controlling insects or arachnids, wherein an active ingredient of the formula I according to one of claims 1 to 20 or a composition comprising this active ingredient is applied to the insect, the arachnid or their environment.
- 47. The use of an active ingredient of the formula I according to one or more of claims 1 to 20 as a herbicide.
- 48. The use of an active ingredient of the formula I according to one or more of claims 1 to 20 as an insecticide or acaricide.
- 49. Seed which has been dressed with an insecticidally or acaricidally active amount of an active ingredient of the formula I according to one or more of claims 1 to 20.

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III. DOCUMENTS CONSI	DERED TO BE RELEVANT		
Category ° Citation	of Document, 11 with indication, where approp	riste, of the relevant passages 12	Relevant to Claim No.13
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ANNEX TO THE INTERNATIONAL SEARCH REPORT ON INTERNATIONAL PATENT APPLICATION NO. 9200452 56863

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.

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